

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YYYY) 02-04-2012		2. REPORT TYPE Final Report		3. DATES COVERED (From - To) May 1, 2006 –October 31, 2011	
4. TITLE AND SUBTITLE THREE-DIMENSIONAL APPROACHES TO ASSEMBLING NEGATIVE INDEX METAMEDIA				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER FA9550-06-1-0279	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) Roberto D. Merlin				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of Michigan, Ann Arbor, MI 48109				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL - OSR - VA - TR - 2012 - 0580	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for Public Release					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT This report describes progress accomplished in the past 5 years in the areas of (i) the design of plasmonic near-field plates for visible operation and corrugated near-field plates for point focusing; (ii) synthesis of gold nanoparticle patterns; (iii) persistent tuning of metamaterials properties; (iv) experimental demonstration of the magnetic moment of a self-assembled cluster of plasmonic nanoparticles; (v) focused-ion-beam synthesis of nanostructure arrays; (vi) search for negative index in chiral molecule composites; (vii) experimental demonstration of negative-index waves in indefinite-permittivity media, (viii) sub-wavelength mid-IR superlensing and (ix) experimental demonstration of negative-index propagation in sub-wavelength plasmonic metamaterials. We also describe advances in the homogenization theory of arbitrary plasmonic and RF metamaterials and the understanding of bulk properties of low-loss periodic negative index structures, as well as results revealing a close relationship between the dynamic magnetic properties of metamaterials and the permittivity of the inclusions, which impose stringent limits to high-frequency magnetism.					
15. SUBJECT TERMS metamaterials, negative refraction, plasmonics, synthesis, focused-ion beam, high-frequency magnetism					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES 19	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			19b. TELEPHONE NUMBER (Include area code)

FINAL REPORT MAY 1, 2006 –OCTOBER 31, 2011

THREE-DIMENSIONAL APPROACHES TO ASSEMBLING NEGATIVE INDEX METAMEDIA

Air Force Office of Scientific Research

Multidisciplinary University Research Initiative

Award Number: FA9550-06-1-0279

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1. RESEARCH OBJECTIVES

The goals of this program were:

- The application and improvement of theoretical models with the goal of identifying conditions for three-dimensional isotropic structures to exhibit true negative refraction, from the millimeter-wave to the infrared range and beyond. Of particular interest were studies to determine the effective permittivity and permeability of metamaterials showing classical, Mie-related resonances as well as mixed magnetic-dielectric aggregates, magnetic semiconductors and chiral molecules, and the exploration of the relationship between intrinsic negative refraction and optical activity, including quantum chemistry calculations to determine spectral parameters.
- The enhancement of existing, and the application of new methods for characterizing the electromagnetic response of systems with negative permittivity and permeability and, in particular, the development of infrared ellipsometry and improved ultrafast optical techniques to study negative-index structures.
- The development of new methods to generate three-dimensional negative-index metamaterials that enhance the capabilities of polymer self-assembly, epitaxy and materials processing techniques. An investigation of organic-matrix structures with metallic and dielectric inclusions and semiconductor-metal hybrids and magnetic-dielectric aggregates as well as a search for molecular systems with large chiral optical anisotropy.

2. ACHIEVEMENTS

Most of the goals delineated in the original proposal were accomplished. This report describes progress accomplished in the past 5 years in the areas of (i) the design of plasmonic near-field plates for visible operation and corrugated near-field plates for point focusing; (ii) synthesis of gold nanoparticle patterns; (iii) persistent tuning of metamaterials properties; (iv) experimental demonstration of the magnetic moment of a self-assembled cluster of plasmonic nanoparticles; (v) focused-ion-beam synthesis of nanostructure arrays; (vi) search for negative index in chiral molecule composites; (vii) experimental demonstration of negative-index waves in indefinite-permittivity media, (viii) sub-wavelength mid-IR superlensing and (ix) experimental demonstration of negative-index propagation in sub-wavelength plasmonic metamaterials. We also describe advances in the homogenization theory of arbitrary plasmonic and RF metamaterials and the understanding of bulk properties of low-loss periodic negative index structures, as well as results revealing a close relationship between the dynamic magnetic properties of metamaterials and the permittivity of the inclusions, which impose stringent limits to high-frequency magnetism.

2.1 Near-Field Focusing Plates

Subwavelength focusing of electromagnetic fields has attracted a great deal of interest in recent years, in particular since the introduction of the superlens by John Pendry in 2000 [1]. A superlens is a slab possessing negative material parameters that can manipulate the near-field and focus electromagnetic waves to resolutions beyond the diffraction limit. In this MURI we proposed a different approach to subwavelength focusing [2], which uses planar surfaces rather than complex volumetric structures to achieve subwavelength focal patterns. These passive surfaces, referred to as near-field plates, excite an electromagnetic field at their surface that mimics the field observed at the exit face of a negative refractive index slab imaging an elemental source. Near field plates are devices that provide focusing well beyond the standard diffraction limit by manipulating the evanescent component of the electromagnetic field. A key issue in the design of a near-field plate is the control of large spatial frequency components. To do so, the right procedures must be found to turn the incident field (usually a plane wave) into a source with a broad angular spectrum, to get large spatial frequencies to resonate and to avoid the presence of unwanted backgrounds [3]. As shown in [4], this requires in turn an amplitude modulation of the optical field exiting the near field plate and an alternating phase between neighboring elements.

During the course of this research, several promising designs have been tested including both metallic and dielectric elements [5]. In [2], a general class of aperture fields (field distributions at the surface of the near-field plate) was proposed that can form a subwavelength focus, and in [3] methods for achieving them were established. In addition, a general procedure was developed for designing near-field plates to achieve desired subwavelength focal patterns. It was recognized that a near-field plate could be implemented as an impedance sheet: a modulated, non-periodic surface reactance consisting of inductive and capacitive elements [3]. The surface reactance sets up a highly oscillatory electromagnetic field that converges to a prescribed subwavelength focus in the near field. The initial implementation of a near-field plate [3] consisted of a non-periodic array of interdigitated capacitors. The plate was designed to focus s-polarized microwave radiation emanating from a cylindrical source to a subwavelength focus with a FWHM of $\lambda/18$. The surface impedance was capacitive given that a capacitive surface supports s-polarized surface waves, and it is these non-radiative waves (evanescent waves in free space) that form the subwavelength focus.

More recently, Grbic and co-workers implemented a new class of near-field plates, consisting of corrugated metallic surfaces that can be directly excited through a waveguide or coaxial feed. The corrugated near-field plates consist of a central aperture in a metallic surface surrounded by non-periodic grooves (corrugations) [6,7]. The central aperture is fed using a waveguide or coaxial feed line, and the surround-

ing grooves act as parasitic radiators which form the subwavelength focal pattern. The grooves can be modeled as short-circuited waveguides, whose impedance can be varied by adjusting their depths.

The development of near-field plates offer distinct advantages over previous designs as they can form unidirectional focal patterns and be directly fed using a waveguide feed or coaxial cable. These features suggest that concentrically corrugated near-field plates can be used as high resolution near-field probes. These devices hold promise for applications in the areas of sensing, near-field optics and wireless power transfer.

2.2 Synthesis of Gold Nanoparticles

Kim's group developed new top-down approaches to fabricate elements for 3D-NIMs. Their studies focused on chemical templates created using self-assembled monolayers to self-assemble nanogold via a charge-charge interaction. Electron-beam lithography (EBL) was used to create these chemical patterns. By varying the exposure dose, it was possible to control the size of the dot for the chemical pattern. Furthermore, by growing a thicker oxide layer on the Si substrate, it was possible to trap the electrons that are backscattered by the Si and generate smaller feature sizes and narrow distribution than with native oxide. Where the resist was exposed during EBL, aminosilane was used to functionalize the substrate with a positive charge. The remaining surface was left as bare silicon-oxide. Previously, they used a wafer that was pretreated with an alkylsilane monolayer to provide a hydrophobic layer to prevent the gold nanoparticles from non-specific binding. However, attempts to reproduce these results with such a wafer were unsuccessful. When a good monolayer of the alkylsilane formed, the contact angle was $\sim 90^\circ$, and the PMMA resist would not spin-coat the wafer. However, when a poor monolayer of the alkylsilane was formed, the contact angle was $\sim 70^\circ$ and the PMMA resist would nicely spin-coat the wafer. The reason that the PMMA would coat the wafer when there was an incomplete monolayer, but not a good monolayer of alkylsilane, was because the surface energy of the good monolayer was too low for the PMMA to wet the surface. For an incomplete monolayer, or the Si-oxide layer (contact angle $\sim 50^\circ$), the surface energy is high enough for the PMMA to coat the wafer.

In other studies, Kim's group explored functionalizing the bare Si-oxide with alkylsilane after depositing the aminosilane. However, this system prevented the nanoparticles from binding to the aminosilane patterned regions. This is believed to occur either because of an interaction between the methoxy-group on the alkylsilane and the amine-group or because the alkylsilane interpenetrated holes in the aminosilane layer, which would then render the pattern hydrophobic due to the alkylsilane (12 carbons long) being longer than the aminosilane (3 carbons long).

During the experiments with the surface coatings, they discovered that the thicker oxide layer on the Si wafer was sufficient enough to prevent non-specific of the negatively charged gold nanoparticles. Thus,

they begun to use samples that are only oxide and patterned aminosilane for our gold nanoparticle assembly. Once the chemical templates are fabricated, they are immersed into an aqueous solution of negatively charged gold nanoparticles, which self-assemble on the positively charged circular chemical patterns. The number of nanoparticles that assemble on the circle depends on the ratio between the diameter of the chemical pattern and the diameter of the nanoparticle. While there is a good control of the number of nanoparticles that assemble, work is still ongoing to achieve uniform loading density on each circular pattern across a very large area. It was found that an immersion time of 20 minutes is sufficient to achieve an equilibrium assembly.

Kim's group made significant progress on a technique called on-rod functionalization, to make Janus rods to produce nanorod pairs in solution. The idea was to pattern rods using EBL, evaporate Au into the patterned area, functionalize the patterned gold with a charge-neutral thiol ligand, lift-off the rods from the substrate, and functionalize the remaining face of the gold rod with a functional thiol ligand having a different character. By creating two batches of such Janus rods, with oppositely charged functional thiol ligands, they managed to obtain self-assembly of rods using a charge-charge interaction in solution. Lift-off was achieved by either ultrasonication of the rod substrate in a good solvent for the ligand or by etching the Ti adhesion layer with HF.

2.3 Persistent Tuning of Metamaterials

Basov's group explored novel approaches to implement frequency-agile metamaterials. Metamaterials which offer dynamically tunable frequency response have been a cornerstone of research geared towards practical device development. Dynamical tuning offers a viable path to overcome limitations associated with the narrow electromagnetic bandwidth inherent to many (or most) metamaterial designs. Basov's group showed how the phase-transition oxide vanadium dioxide can be used to enable infrared dynamic-tuning of an array of split-ring-resonators (SRRs) using the hybrid-metamaterial architecture [8]. This novel approach pioneered by the UCSD team is now employed by many research groups around the globe. Basov and co-workers also demonstrated *persistent* switching of resistance of VO₂ [9] and capitalized on this effect to demonstrate memory metamaterials [10].

Using a thin-film device, Basov's group demonstrated electrical voltage-based control of the phase-transition of VO₂. They found that properties of the VO₂ film associated with the phase-transition, such as the DC resistivity, can be advanced in a 'ratchet-like' manner using brief transient voltage pulses as a stimulus [9]. In all previously demonstrated dynamic tuning schemes, continuous application of external stimulus was required to maintain tuning. Leveraging the new ratchet effect in the VO₂ phase-transition, several schemes for enabling *persistent* tuning in a metamaterial could be outlined. Incorporation of persistent tuning in metamaterials would greatly advance the utility of practical devices, such as frequency-

agile antenna arrays and infrared electro-optic devices, as it would not require incessant external stimulus. Hybrid SRR-VO₂ metamaterial structures were fabricated and memory switching of the resonant behavior of this structure was demonstrated [10]. A by-product of this work was the demonstration of new effect of memory capacitance that is of high interest in the context of numerous device applications. Experiments utilizing the near-field nanoscope being developed in Basov's group are currently being employed to probe the physics behind this unique property of VO₂.

2.4 FIB-Synthesized Nanostructure Arrays for Negative Index Metamaterials

Goldman's group developed a new method for the fabrication of materials with novel physical properties not found in nature, namely, focused-ion-beam (FIB) assisted molecular beam epitaxy (MBE) of highly-ordered Ga nanodroplet arrays within semiconductors. Significant progress was also achieved towards the fabrication of highly-ordered 3D Ga nanodroplet arrays using a combination of FIB patterning of Ga droplets followed by MBE overgrowth of GaAs layers. It has been predicted that ordered arrays of plasmonic nanospheres in a matrix will lead to simultaneously negative permittivity and permeability, paving the way to low loss negative-index metamaterials (NIMs) [11]. Among various metallic nanostructures with plasmonic behavior, Ga droplets show surface plasmon resonance up to 3.3 eV for 10 nm-sized nanodroplets. Therefore, arrays of Ga nanodroplets within semiconductors are promising candidates for NIMs operating up to visible frequencies [12]. On III-V semiconductor surfaces, nanometer-sized metallic amorphous liquids (i.e. droplets) or crystalline solids (i.e. dots), mostly consisting of group III elements, often form during thermal annealing, exposure to a group III element, and/or ion irradiation. In the case of focused-ion beam (FIB) irradiation of III-V semiconductor surfaces, group V elements are preferentially sputtered, forming a group III-rich ion-milled region. With continued irradiation beyond a threshold ion dose, group III-rich droplets or dots were observed [13].

The influence of interdroplet spacing and droplet diameter on the plasmon resonance energy were studied in considerable detail. The plasmon resonances for arrays with small droplet diameter (~ 120 nm) and interdroplet spacing (~ 400 nm) was compared with those of larger droplet diameter (~ 200 nm) and interdroplet spacing (~ 900 nm). Transmittance and reflectance spectra in the range 400-1000 nm were measured. The resulting extinction spectra reveal plasmon resonances at ~ 520 and ~ 790 nm, respectively. The data shows the expected red-shift of the resonances as the interdroplet spacing and droplet diameter increase.

2.5 Search for NIMs in Isotropic Media of Chiral Molecules (CNIMs)

Work was aimed at assessing the properties of thin isotropic films of chiral molecules or assemblies possessing spectral regions of negative refractive index, CNIMs. A comprehensive theoretical report was produced that connects the optical properties of a homogeneous, three-dimensional, CNIMs to the quan-

tum mechanical properties of individual chiral molecules constituting or imbedded in this medium. CNIMS exist under the requirement that the unpolarized real refractive index of a material is closer to zero than half the magnitude of the circular birefringence (CB), the difference in the refractive index for left versus right circularly polarized radiation, $\Delta n = n_L - n_R$. CNIMs represent a unique class of NIMs since both the permittivity and permeability may be positive and the permeability may be near unity. Quantum chemistry calculations using density functional theory (DFT) were extended and evaluated for chiral helicenes, cyclacenes (short segments of chiral carbon nanotubes) and chiral fullerenes, the simplest member of which is a closed, near-spherical 76-carbon-atom structure. The calculations performed point to the need for even stronger electronic absorption, as well as large CB, hence our investigations of chiral cyclacenes and fullerenes. This work uncovered the presence of low-lying infrared excited electronic states in selected cyclacenes and fullerenes. These infrared electronic excited states couple with vibrational transitions to produce extremely intense narrow vibrational absorption and vibrational CB (VCB) bands. We have thereby shown that these molecules are excellent candidates for fabricating CNIM thin films. For the typically narrow wavelengths over which negative refraction exists, the CNIM is a one-element circular polarization beamsplitter or circular polarizer, thereby forming an entirely new class of photonic polarization device.

2.6 Extraction of constitutive parameters

Shvet's group embarked on a new research direction aimed at developing a new extraction procedure for all 36 constitutive parameters of an arbitrary metamaterial, both on and off the propagation shell. Such extracted parameters can be used for rapid simulation of embedded antennas into an arbitrary metamaterial. The homogenization procedure, referred to as current-driven homogenization (CDH) fully takes into account spatial dispersion. This CDH extraction procedure is more fundamental than the traditional extraction using the conventional S-matrix (scattering matrix) approach for several reasons. First, there is a number of applications such as, for example, radiation by embedded antennas of complex spatial shape) that require the full knowledge of the constitutive parameters such as $\varepsilon(\omega, k)$, $\mu(\omega, k)$, $\varepsilon(\omega, k)$, and $\xi(\omega, k)$ for arbitrary (and mutually independent) (ω, k) sets. Second, the conventional $\varepsilon(\omega, k(\omega))$ extracted on the propagation shell can exhibit rather unphysical properties such as anti-resonances, etc. By extracting the constitutive parameters at the Gamma-point (e.g., $\varepsilon(\omega, k = 0)$), as opposed to the more conventional $\varepsilon(\omega, k(\omega))$ on the propagation shell), they demonstrated that some of these unphysical features can be removed and, therefore, attributed to spatial dispersion.

One example of such an extraction for a negative-index metamaterial with a unit cell comprised of a high-index dielectric surrounded by a negative-permittivity shell. The extracted dielectric permittivity and magnetic permeability show strong a magnetic resonance which results in an electric anti-resonance at the same frequency, which disappears for $\varepsilon(\vec{k} = 0, \omega)$. Details of this procedure are published in [14].

2.7 Self-Assembled Plasmonic Nanoparticle Clusters

This work was done in collaboration with the groups of Capasso at Harvard and that of Halas-Nordlander at Rice. The goal was to develop a new bottom-up fabrication technique for colloidal nanoparticles exhibiting magnetic resonances. Computational techniques for modeling the response of such nanoclusters were developed in the Texas group of Shvets, and were crucial for interpreting the experimental results, published in [15].

The self-assembly of colloids is an alternative to top-down processing that enables the fabrication of nanostructures. We showed that self-assembled clusters of metal-dielectric spheres are the basis for nanophotonic structures. By tailoring the number and position of spheres in close-packed clusters, plasmon modes exhibiting strong magnetic and Fano-like resonances emerge. The use of identical spheres simplifies cluster assembly and facilitates the fabrication of highly symmetric structures. Dielectric spacers are used to tailor the interparticle spacing in these clusters to be approximately 2 nanometers. These types of chemically synthesized nanoparticle clusters can be generalized to other two- and three-dimensional structures and can serve as building blocks for new metamaterials. Experiment was guided by numerical simulations. First, we have identified the electrostatic resonances that give rise to magnetic and electric dipole responses. The simulations revealed a very strong magnetic resonance at the low (IR) frequency. This indicated to experimentalists the frequency range where magnetic activity can be revealed. While magnetic resonance is generally obscured by the electric dipole resonance, it is clearly revealed by the cross-polarized measurements. In addition to magnetically-active trimers, more complicated clusters such as heptamers were also assembled and investigated using dark field microscopy.

2.8 Generalized phase matching for lossy periodic photonic structures

Negative refraction in lossless dielectric photonic crystals has been well understood using a phase matching condition across an interface of periodicity d , $\mathbf{k}_t^B = \mathbf{k}_t^m + m\mathbf{G}$. Here, $|\mathbf{G}| = 2\pi/d$ is the magnitude of the interface reciprocal lattice vector, m is an integer for denoting different diffraction orders, and $\mathbf{k}^B$ and $\mathbf{k}^m$ are *real* wave vectors of a Bloch wave and a plane wave, respectively. In the more general case of lossy periodic media, however, Bloch waves acquire an *imaginary* component of the wave vector. Diffraction of complex Bloch waves by periodic photonic structures has not been generally treated. Negative

refraction has also been observed in negative index metamaterials (NIMs), which are typically periodic lossy metal-dielectric composites. Although complex band structure of metamaterials has been routinely calculated, negative refraction by composite NIM prisms is generally interpreted as the behavior of a homogeneous NIM with effective permittivity and permeability.

In studies pursued in Forrest's group, NIM refraction experiments were treated using a generalized phase matching condition with a complex transverse wave vector for periodic media [16]. The diffraction of a complex Bloch wave propagating within a composite prism was described, and shows that the detected light is an inhomogeneous plane wave due to losses. They have also shown that the negative refractive behavior of lossless dielectric photonic crystals and lossy metal-dielectric periodic NIMs can be given a unified explanation, and discussed its implications for the local homogeneous model of NIMs as well as the possible existence of a minimum unit cell size of optical NIMs.

2.9 Phase advance by a subwavelength near-infrared NIM

In a collaboration with the groups of Gennady Shvets and Xiaoqin Li at the University of Texas, the group of Forrest developed novel interferometric measurements of the phase advance of light through an optical negative index metamaterial slab. Such measurements have rarely been reported in the literature. In the few such reports available, the relationship between the measured phase advance through samples consisting of only a single layer of unit cells and that expected for transmission through hypothetical multi-layer bulk NIMs remains unclear. Moreover, most optical NIMs have a unit cell size approaching half of the wavelength of interest in at least one dimension. This large unit cell makes their description as effective media problematic. They were able to measure [17] the relative phase shift between *s*- and *p*-polarized light introduced by a single layer of a subwavelength near-infrared negative index metamaterial structure, which is consistent with theoretical predictions based on the geometrical dimensions of the structure. Numerical scattering simulations further suggest that the negative phase advance for the *p*-polarized light through the single-layer sample is similar to the negative phase advance per unit cell expected to be exhibited by a bulk material comprising multiple layers of the structure.

2.10 Limits to High-Frequency Magnetism

Homogeneous composites, or metamaterials, made of dielectric or metallic particles are known to show magnetic properties that contradict arguments by Landau and Lifshitz [*Electrodynamics of Continuous Media* (Pergamon Press, Oxford, 1960), p. 251] indicating that the magnetization and, thus, the permeability loses its meaning at relatively low frequencies. Merlin [18] showed that these arguments do not apply to composites made of substances with $\kappa_s = \text{Im}\sqrt{\epsilon_s} \gg \lambda/\ell$ or $n_s = \text{Re}\sqrt{\epsilon_s} \sim \lambda/\ell$ (ϵ_s and ℓ are the com-

plex permittivity and the characteristic length of the particles, κ_s and n_s are, respectively, the extinction coefficient and the refractive index, and $\lambda \gg \ell$ is the vacuum wavelength). The general analysis is supported by studies of split-rings, one of the most common constituents of electromagnetic metamaterials, and spherical inclusions. An analytical solution was given to the problem of scattering by a small and thin split ring of arbitrary permittivity. Results reveal a close relationship between ϵ_s and the dynamic magnetic properties of metamaterials. For $|\sqrt{\epsilon_s}| \ll \lambda/a$ (a is the ring cross-sectional radius), the composites exhibit very weak magnetic activity, consistent with the Landau-Lifshitz argument and similar to that of molecular crystals. In contrast, large values of the permittivity lead to strong diamagnetic or paramagnetic behavior characterized by susceptibilities whose magnitude is significantly larger than that of natural substances.

The double constraint $\kappa_s \gg \lambda/\ell \gg 1$ (or, $n_s \sim \lambda/\ell \gg 1$ if $\kappa_s \ll n_s$) poses severe limitations for attaining magnetism at arbitrarily high frequencies. Because they have a large extinction coefficient, metals are to be favored at optical frequencies. Since $\epsilon_s \approx -\omega_p^2/\omega^2$ for $\omega\tau \gg 1$, the constraint becomes $\lambda \gg \ell \gg \lambda_p$. The measured values of the permittivity for noble metals indicate that magnetism can coexist with the effective-medium condition for frequencies up to $\sim 1.5 \times 10^{14}$ Hz ($\lambda \sim 2.5 \mu\text{m}$).

2.11 Magneto-Electric Coupling in Metamaterials

In studies pursued in the Duke group of David Smith, magnetoelectric coupling in negative index media was investigated. In the past, analytical formulas were found for metamaterials that have either magnetic or electric resonances, but not both. These analytical formulas, though simple in form, exhibit excellent agreement with numerical retrievals and can be used as the basis for more intelligent optimization, design or interpolation schemes. When both electric and magnetic resonances exist in a unit cell, magnetoelectric coupling can occur between the elements, and the previously derived formulas are incorrect. In fact, numerical retrievals will typically fail in the presence of magnetoelectric coupling, either within the same unit cell or between unit cells. The effective refractive index can be calculated analytically for a cell with both electric and magnetic resonators, and leads to analytical expression that can be easily tested. As a function of the coupling strength, very discernable artifacts appear in the effective index. Results show that numerical simulations on a cell with electric and magnetic resonators yields exactly the same curves as the analytical expressions.

2.12 Metamaterials Parametric Oscillators

Optical parametric generators (OPG) and oscillators (OPO) have a wide applications spectrum, ranging from broadband wavelength conversion to entangled photon generation. OPGs exhibit high gain and a

possibility of a broadband generation by tuning the phase-matching conditions for the parametric down-conversion process. They also have a relatively simple set-up compared to that of an OPO, as no external cavity and alignment is required. However, OPGs generally have a high threshold level such that strong laser pump pulses are required. Also, as a consequence of having no external cavity, less control over the emitted signal and idler pulses is available. These limitations can be overcome by the OPO design, which has a lower threshold operation and allows a fine control over the frequency of the emitted pulses by cavity resonance. However, with regular materials, the design requires a fine tuning of the phase-matching conditions to the resonance wavelength of the cavity. This also leads to a single-wavelength operation of the OPO. We suggested a metamaterial-based design of an OPO that overcomes these limitations.

In the presence of nonlinear susceptibility, the wave equation at the pump and harmonic frequencies are coupled. It is possible to solve the coupled system analytically in an infinite medium, but not for a slab of material. However, if the combination of the nonlinearity and the incident power is small, one can suppose that the nonlinearity does not significantly alter the distribution of the electric field at the pump frequency. This is called the non-depleted pump approximation. Using the non-depleted pump approximation one can: (i) calculate the distribution of the electric field at the pump frequency using a linear transfer matrix approach; (ii) use the electric field in the slab and the nonlinear susceptibility to calculate the component of the nonlinear polarization at the harmonic frequency; (iii) couple that nonlinear polarization to an electric field at the harmonic frequency and (iv) propagate that electric field to the input and output medium using a linear transfer matrix approach at the harmonic frequency. Using such an approach, Smith and co-workers showed that this process can be reversed and that knowing the applied field at the pump frequency, the amplitude of the generated harmonic, and the linear electromagnetic properties at both the pump and the harmonic frequencies, it is possible to determine the nonlinear susceptibility. They have applied the nonlinear retrieval method to the simulation of second harmonic generation in a simplified cut wire sample. The simulated unit cell consists of a bar of copper centered in a unit cell of nonlinear dielectric. The copper bar is slightly shorter than the unit cell, leaving a small gap between adjacent copper bars, and concentrating the electric field. Using Comsol, they have simulated the generation of harmonics of this sample with 1, 2, 3, or 4 unit cell thick. Then, they used the retrieval method to determine the effective nonlinear susceptibility. The results indicate that the nonlinear susceptibility is 16 times higher than that of the nonlinear dielectric used in the unit cell (this can be related to the concentration of the electric field in the gap). There is a small distribution of the values determined for the reflected or transmitted second harmonic, as well as a function of the number of unit cells. This can be related to the homogenization of the material.

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4. PERSONNEL SUPPORTED

co-PI	GRADUATE STUDENTS	POSTDOCTORAL FELLOWS
Dimitri Basov	G. Andreev	T. Driscoll
Stephen R. Forrest	X. Zhang	
Rachel S. Goldman	M. Kang and Jia-Hung Wu	
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Jinsang Kim	R. Nidetz	
Roberto Merlin	A. Bianchini & I. Vugmeister	
Lawrence A. Nafie	R. A. Lombardi	T. B. Freedman
Gennady Shvets	A. Kanikaev B. Neuner	Y. Avitzour & D. Korobkin
David R. Smith	D. Huang, R. Liu & J. Allen	E. Poutrina & S. Larouche

5. PUBLICATIONS (2006-2011)

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6. PATENT DISCLOSURES

1. R. Liu, T. J. Cui, J. Gollub, Q. Cheng and D. R. Smith: "Metamaterials for Surfaces and Waveguides." U. S. Patent Application No. 20100156573.
2. R. Merlin and A. Grbic: "Apparatus for Subwavelength Near-Field Focusing of Electromagnetic Waves." U. S. Patent Application No. 60/938,858 (pending).
3. D. R. Smith, "Broadband Metamaterial Apparatus, Methods" PCT/US2010/021240

7. HONORS AND AWARDS

Dimitri Basov is a Fellow of the American Physical Society and received the 2004 Genzel Prize, awarded to a young scientist for exceptional contributions to the field of condensed-matter spectroscopy.

Steve Forrest is a Fellow of IEEE and the Optical Society of America. He received the Powell Foundation New Investigator Award, University of Southern California, the IPO Distinguished Inventor (1998), the 1998 Thomas Alva Edison Award for innovation in emerging technology, the 1999 Materials Research Society Medal for pioneering contributions to the growth and optoelectronic applications of organic semiconductor thin films, the 2001 IEEE/LEOS W. Streifer Award for contributions to the development of APDs and pin detectors for long wavelength optical communication systems, the 2006 Jan Rajchman Prize of the Society Information Display for insights into exciton generation and diffusion that led to the

discovery of phosphorescent OLEDs, quadrupling efficiency, and the 2007 IEEE Daniel Noble Prize for innovations in organic light emitting devices. Other honors include IEEE Distinguished Lecturer (1996-1997) and William Mong Distinguished Lecturer (University of Hong Kong). Forrest was elected to the National Academy of Engineering in 2003.

Rachel Goldman was the recipient of a Radcliffe Faculty Fellowship (2005-2006), from the Radcliffe Institute of Advanced Study, Harvard University, and received the 2004 Ted Kennedy Family Team Excellence Award, of the University of Michigan College of Engineering and the 2002 Peter Mark Memorial Award of the American Vacuum Society.

Lawrence Nafie received the 2001 OSA Meggers Award and the 2001 Bomem Michelson Award, the Chancellor's Citation for Exceptional Academic Achievement, Syracuse University (1998) and the 1981 Coblentz Award for outstanding contributions to molecular spectroscopy.

Roberto Merlin is a Fellow of the American Physical Society, the Optical Society of America, the von Humboldt Foundation and the John Simon Guggenheim Memorial Foundation. Other honors include the 2006 Frank Isakson Prize of the American Physical Society for Optical Effects in Solids and Lannin Lecturer (2002) at the Department of Physics, Pennsylvania State University.

Gennady Shvets received a Presidential Early Career Award for Scientists and Engineers (2000).

David R. Smith was selected as a member of The Electromagnetics Academy in 2001. He was a co-recipient of the 2004 Descartes Research Prize awarded by the European Union and the 2005 Stansell Research Award from the Pratt School of Engineering.