Negative Refraction in the Optical Range and Left-Handed Photonics

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Section I. Foreword

Optical negative-index metamaterials are one of the newest and perhaps one of the most exciting areas of optical science and physics. This project, focused on optical negative refraction and left-handed photonics, has provided a number of novel and useful results. This document briefly describes our studies on negative refraction in the optical range and left-handed photonics. We summarize the most important project results in Section III. Our work has provided several truly remarkable and novel results, including:

- The first demonstration of an optical negative-index metamaterial,
- The first demonstration of artificial magnetism across the entire visible part of the spectrum,
- The first design for an optical cloak,
- Significant advances in the fundamentals of nonlinear optics of negative-index metamaterials,
- Sophisticated modeling and designs for tunable NIMs, and
- Proposals for planar hyperlens designs and other transformation-optics devices.

Section II. Statement of the Problem Studied

In this project, we were concerned primarily with studying the physical phenomena, devices and designs related to negative refraction in the optical frequency range. We used theoretical work, computer simulations, and experimental studies to further the understanding of negative refraction in the high sought-after optical range. Our work has brought a number of significant advances, including experimental demonstrations of negative refraction well into the visible range to advanced designs in hyperlens and cloaking devices. The physics and repercussions of negative refraction in nonlinear optical systems has also been considered in our work, with the goal of understanding this newly opened and extremely exciting field of optics research.

Section III. Summary of the Most Important Results

The summary of the important results from this project are presented in several sub-sections in order to present the work and our results in a clear and understandable way.

Metamagnetics: Non-unity permeability at optical frequencies

The first group of results is related to our efforts to create optical magnetic responses in metamaterials. A magnetic response is required for low-loss negative-index metamaterials. For optical metamaterials, this is a challenging requirement to meet because no natural materials exhibit a magnetic response in the optical frequency range.

During this project we have experimentally demonstrated experimentally a material with a negative permeability in the red part of the visible spectrum [1]. Using a non-periodic array of pairs of thin silver strips, we fabricated two samples with different strip thicknesses. Optical measurements of the samples confirm our initial design projections by showing the real part of permeability to be about −1 for the sample with thinner strips and −1.7 for the sample with thicker strips at wavelengths of 770 nm and 725 nm, respectively. This shows that we have the ability to create a magnetic response, a permeability value different than unity, within the visible frequency range.

We have also shown that it is possible to create magnetic responses across the whole visible spectrum [2]. This metamagnetic material, fabricated using a family of coupled nanostrips with varying dimensions, exhibits optical magnetic responses through the entire visible spectrum from red to blue. We refer to such a phenomenon as rainbow magnetism. The experimental and analytical studies of such structures provide us with a universal building block and a general recipe for producing controllable optical magnetism for various practical implementations.

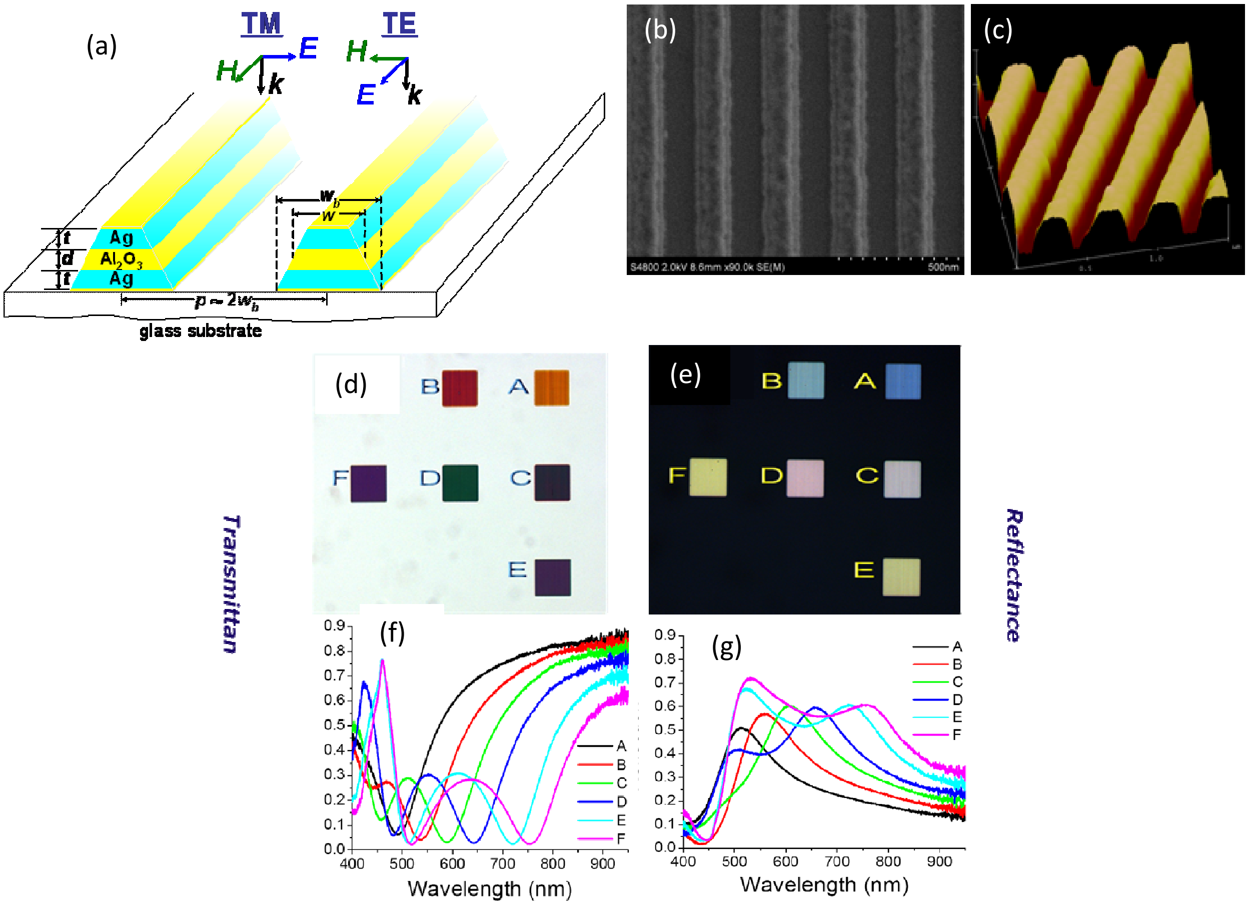


Figure 1: Metamagnetics in the visible frequency range. Samples of coupled silver strips separated by alumina are fabricated on glass substrates (a). Representative SEM and AFM images show the grating pattern of the fabricated samples (b, c). The parameters of the gratings are varied to produce samples labeled A-F (d, e) whose magnetic responses are observed across the visible frequency range under appropriately polarized incident light (f, g).

optical range. Artificially engineered metamaterials are now demonstrating unprecedented electromagnetic properties that cannot be obtained with naturally occurring materials. In particular, they provide a route to creating materials that possess a negative refractive index and offer exciting new prospects for manipulating light. Indeed, recent progress made in creating nanostructured metamaterials with a negative index at optical wavelengths has been rapid and exciting; one of our results of this project is a comprehensive review of these metamaterials and a discussion of some of the devices that could result from these new materials [3]. We also presented simulation results of a design for negative-index materials that uses magnetic resonators to provide negative permeability and a metal film for negative permittivity [4]. In this work, we also introduced the possibility of using semicontinuous metal films to achieve better manufacturability and enhanced impedance matching in the metamaterials.

Our first experimental results in this sub-section occurred at a very important telecommunications wavelength [5]. An array of pairs of parallel gold nanorods is shown to have a negative refractive index at the optical communication wavelength of 1.5 μm. This effect results from the plasmon resonance in the pairs of nanorods for both the electric and magnetic components of light. The refractive index is retrieved from the direct phase and amplitude measurements for transmission and reflection, which are all in excellent agreement with our finite difference time domain simulations. The refraction critically depends on the phase of the transmitted wave, which emphasizes the importance of phase measurements in finding $n' \approx -0.3$.

In another result at the same wavelength, we demonstrated a negative refractive index that reached −2 using specially designed metal-dielectric composites [6]. Specifically, we showed that arrays of single and paired nanorods can provide such negative refraction. For pairs of metal rods, a negative refractive index has been observed at 1.5 μm. The inverted structure of paired voids in metal films may also exhibit a negative refractive index. A similar effect can be accomplished with metal strips in which the refractive index can reach −2. The refractive index retrieval procedure and the critical role of light phases in determining the refractive index is discussed.

We even observed dual-band negative-index behavior (showing both a negative effective permeability and a negative effective permittivity) from a metamaterial design in the near-infrared frequency range [7]. The performance of these materials has been analyzed using a full-wave numerical electromagnetic scattering method. Negative effective permittivity is provided by a thin layer of a metallic film. Negative effective permeabilities are created in two distinct frequency bands by magnetic resonators of different dimensions.

Further experiments in this project have shown negative-refractive-index behavior at wavelengths as short as 770 nm, providing additional experimental verification of optical negative-index materials. Our comprehensive studies on a periodic array of gold nanorod pairs demonstrate its unique optical properties, including a negative refractive index in the optical range [8]. In particular, we provided an experimental demonstration of a dual-band negative index metamaterial in the visible frequency range [9]. The sample is double-negative (showing both a negative effective permeability and a negative effective permittivity) for wavelengths between 799 and 818 nm of linearly polarized light with a real part of refractive index of about −1.0 at 813 nm; the ratio −Re(n)/Im(n) is close to 1.3 at that wavelength. For an orthogonal polarization, the same sample also exhibits a negative refractive index in the visible (at 772 nm). The spectroscopic measurements of the material are in good agreement with the results obtained from a finite-element electromagnetic solver for the actual geometry of the fabricated sample at both polarizations.

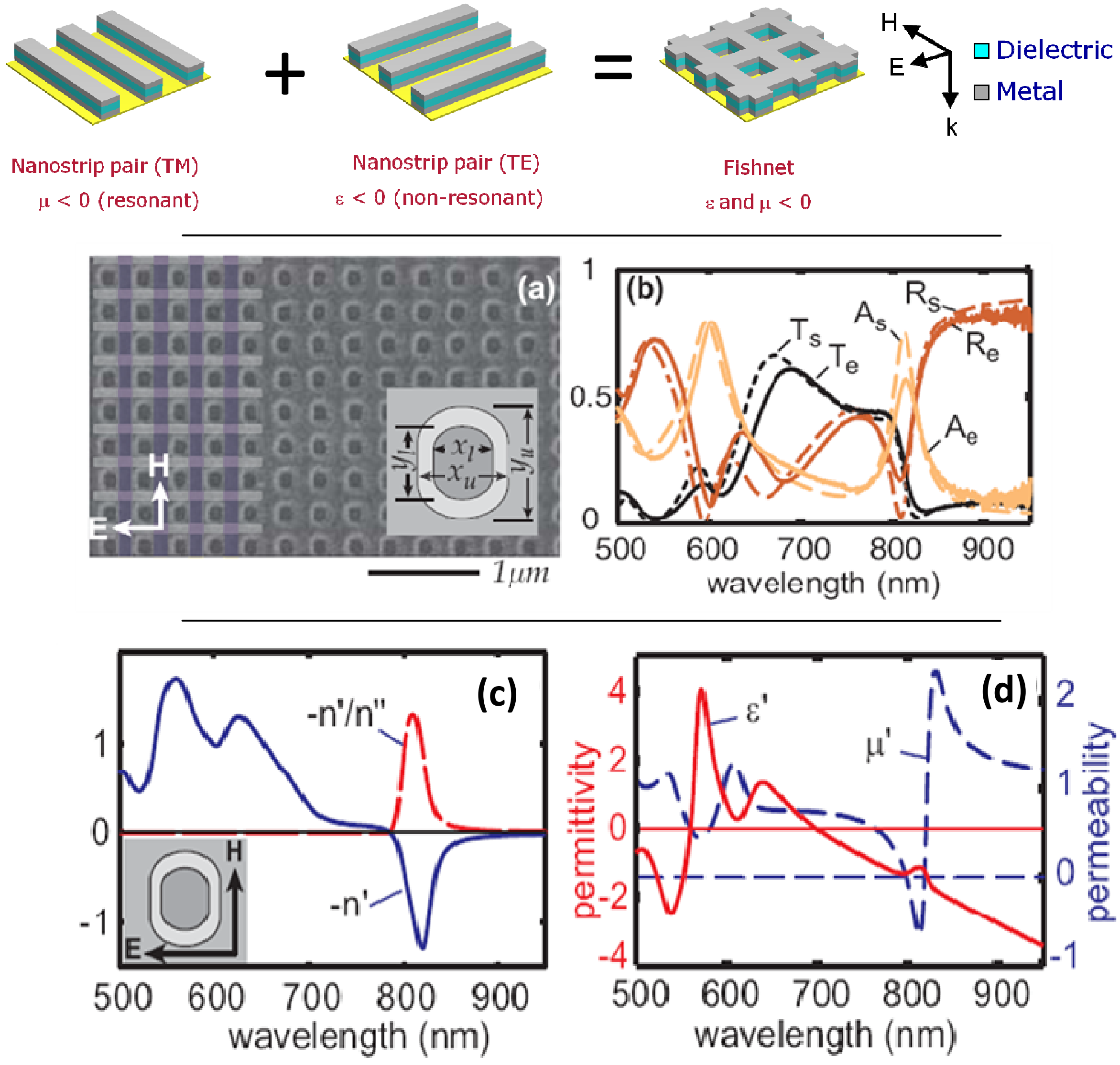


Figure 2: Negative-index fishnet metamaterials. The uppermost panel shows the general recipe for creating a fishnet negative-index metamaterial. Middle panel: (a) SEM image of the negative-index fishnet sample. The inset in (a) depicts the unit cell of the simulated structure. (b) A comparison of the experimental transmission (Te), reflection (Re) and absorption (Ae) spectra of the sample with simulated results (Ts, Rs, and As) at the primary linear polarization as shown in (a). Lower panel: The sample in double-negative, negative-index regime. The primary polarization of light is used in modeling as shown in the inset. (a) The real part of the effective refractive index and figure of merit. (b) The real part of the effective permeability (μ') and permittivity (ε') are both negative from 799 to 818 nm.

We have also investigated the possibility of tuning the index of refraction in a metamaterial, giving a variable effective refractive index that can be negative, zero or positive [10]. Near-infrared metamaterials that possess a reconfigurable index of refraction from negative through zero to positive values were developed. The reconfigurability was achieved by cladding thin layers of liquid crystal both as a superstrate and a substrate on an established negative-index metamaterial and adjusting the permittivity of the liquid crystal. Numerical results showed that the index of refraction for the proposed structure can be changed over the range from −1 to +1.8 by tuning the liquid crystal permittivity from 2 to 6 at a wavelength of 1.4 μm. A full-wave analysis technique based on the finite element-boundary integral method was developed and used to rigorously treat the scattering from periodically structured metamaterials incorporating anisotropic liquid crystals and dispersive materials [11]. Reconfiguration of the negative-index metamaterials was achieved by controlling the magnetic resonance via tuning permittivity of the embedded anisotropic liquid crystals. Our numerical results showed that the refractive index of the metamaterials can be reconfigured by tuning the director orientation of anisotropic liquid crystals or by using temperature-dependent liquid crystals. The design configurations and their characteristics in the near- and the mid-infrared ranges were presented.

Nonlinear optics in negative-index materials

Nonlinear optics is a field of optical science that contains many truly fascinating phenomena, from second-harmonic generation to self-focusing and even soliton propagation. The extraordinary properties of second-harmonic generation in negative-index metamaterials, where the Poynting vector and the wavevector have opposite directions, were investigated within this project [12]. The opposite directions of the wave vector and the Poynting vector in negative-index materials results in a ”backward” phase-matching condition, causing significant changes in the Manley-Rowe relations and spatial distributions of the coupled field intensities. Further studies included optical parametric amplification in left-handed materials [13,14], where we have shown that absorption in negative-index metamaterials can be compensated by backward optical parametric amplification. The optical parametric amplification process can be controlled by the auxiliary electromagnetic field and enables transparency, amplification, and oscillation with no cavity in strongly absorbing negative-index metamaterials. The feasibility of generating entangled pairs of left- and right-handed counter-propagating photons was also discussed in this work.

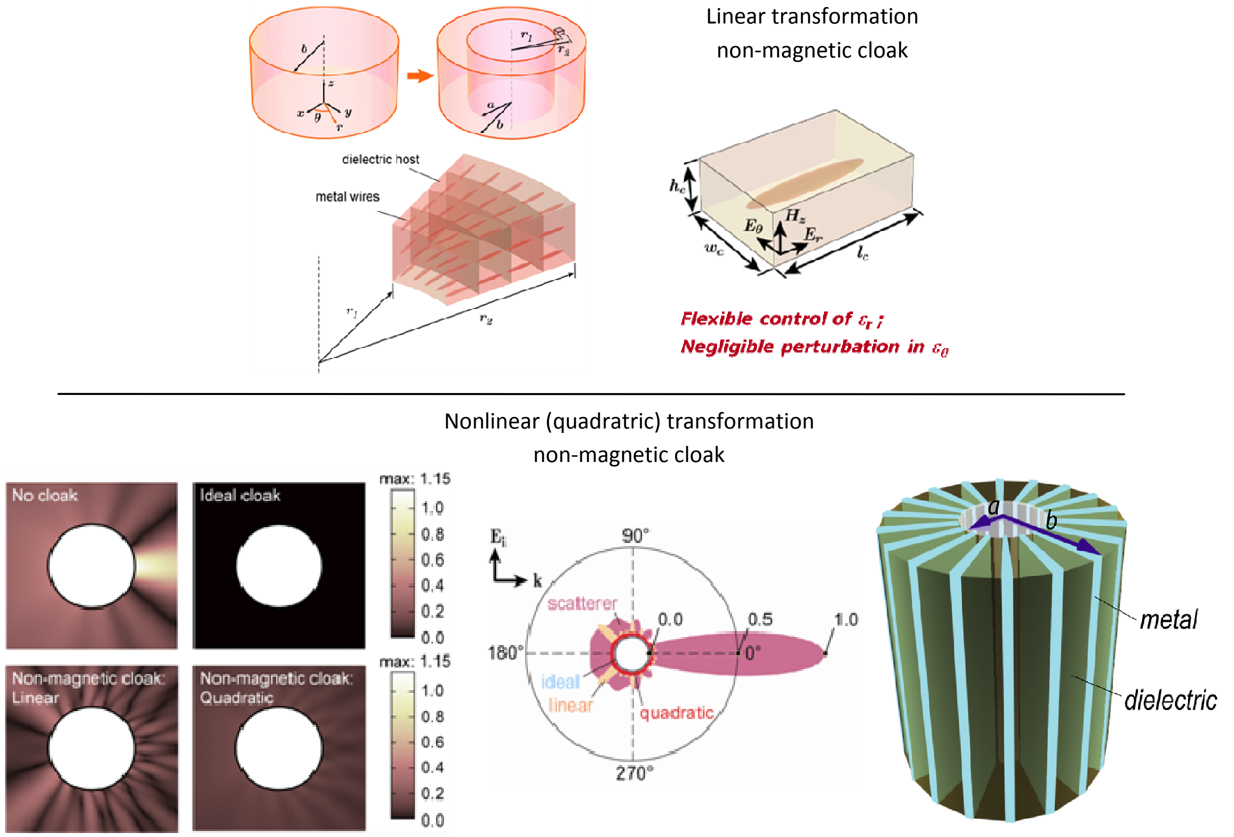
New physics and new devices

In this sub-section, we discuss results related to new phenomena, new methods of analysis and new devices in negative-index metamaterials. Our experimental studies and computer simulation results have shown some very interesting and useful properties of these systems, including the possibilities of nonmagnetic cloaking, hyperlens designs, and other devices.

One of the most exciting possibilities arising from metamaterials is that of optical cloaking. In this project, we have developed and simulated numerically a non-magnetic cloak with minimized scattering [15]. In an electromagnetic cloak based on a transformation approach, reduced sets of material properties are generally favored due to their easier implementation in reality, although a seemingly inevitable drawback of undesired scattering exists in such cloaks. We suggested the use of high-order transformations to create smooth moduli at the outer boundary of the cloak, therefore completely eliminating the detrimental scattering within the limit of geometric optics. We applied this scheme to a nonmagnetic cylindrical cloak and demonstrated that the scattered field is reduced substantially in a cloak with an optimal quadratic transformation as compared to its linear counterpart.

A critical issue in the development of metamaterials and nanoscale plasmonics in general is an understanding of the material properties of the constituent materials that make up the composite structure. Silver, the best plasmonic material due to its relatively low loss, can exhibit different material properties when the metal structures are in the nanoscale [16]. We have found that the Ag permittivity (dielectric function) in coupled strips is different from the bulk value; we have studied this phenomenon for strips of various dimensions and surface roughness. Arrays of such paired strips exhibit the properties of a metamagnetic material, an important precursor for a negative refractive index in the optical range. The surface roughness does not affect the real part of the Ag dielectric function, although it does increase the loss at the plasmon resonances of the coupled strips.

Recent advances in metamaterial research have provided us a blueprint for realistic cloaking capabilities, and it is crucial to develop practical designs to convert concepts into real-life devices. In this project, we have studied a number of designs for optical cloaks [17]. Two such structures for optical cloaking are based on high-order transformations for TM and TE polarizations. The high-order transformations allow more flexibility in the design constraints, producing more realistic or more suitable distributions of material parameters within the cloak structure. These designs are possible for visible and infrared wavelengths. This critical development builds upon our previous work on nonmagnetic cloak designs and high-order transformations.


Linear transformation non-magnetic cloak

Flexible control of ε_r ;
Negligible perturbation in ε_θ

Nonlinear (quadratric) transformation non-magnetic cloak

Figure 3: Non-magnetic optical cloaking designs. The linear-transformation cloak (top panel) shows good cloaking performance, but is plagued by scattering problems. The nonlinear-transformation cloak (bottom panel) avoids the scattering issues and still provides excellent cloaking performance.

some of the best theoreticians and researchers in this field, in this project we have presented a design for a cylindrical hyperlens made of a layered binary material [18]. The design approach uses an improved effective medium theory to take account of radius-dependent effects due to curvature of material interfaces resulting in nonperiodically distributed thicknesses of the lens layers. The performance of this lens is compared versus the designs with periodically thick layers, which we showed in earlier papers. Detailed quantitative results analyzed for the lenses with the same number and starting order of layers prove better functioning of the lens designed with this approach.

In exploring hyperlens designs, we turned our attention to the field of transformation optics. One of the most powerful results of this project is the further understanding and development of transformation-optics approaches for designing metamaterials. Using transformation optics, researchers can actually engineer optical space, molding the flow of light in an almost arbitrary way. One of our specific results combined conceptual studies and numerical simulations for imaging devices that both transform a near-field pattern into magnified far-zone images and are based on a high-order spatial transformation in cylindrical domains [19]. A lens translating a near-field pattern from an almost circular input boundary onto a magnified, far-field image at a flat output boundary was considered. The lens was made of a metamaterial with anisotropic permittivity and permeability both depending on a single "scaling" parameter of the transformation. Open designs of the lens with a truncated body (3/4-body and 1/4-body lenses) were suggested and analyzed. We showed that the ideal, full lens and the 3/4-body lens produce identical images. Numerical simulations of 1/4-body designs indicate that further truncation of the lens could limit its performance. A light concentrator "focusing" far-zone fields into a nanometer-scale area was also considered.

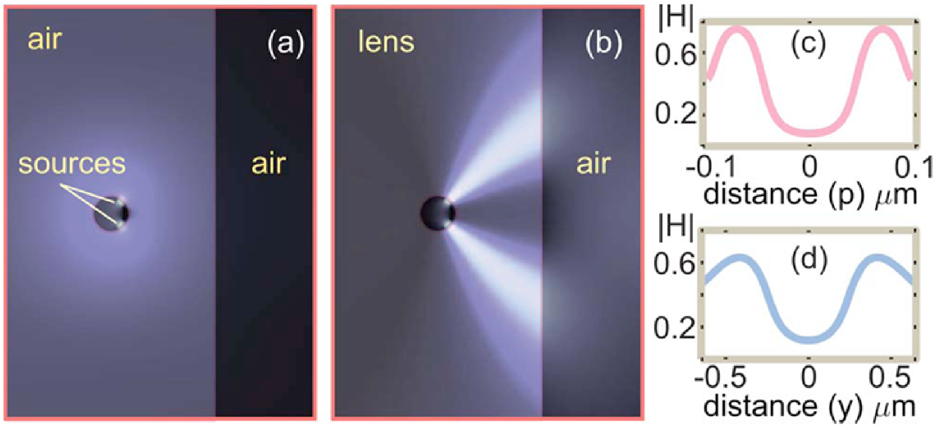


Figure 4: Simulation of a closed, flat hyperlens design showing the near-to-far-field projection capabilities of the lens. (a) Magnetic field intensity generated by two coherent test sources in air. (b) Magnetic field intensity generated by the pair of test sources inside and just outside the lens. Shaded areas indicate the lens cross section in (a) and (b). (c) H-field magnitude generated by the sources along the curvilinear (input) surface of the lens. (d) H-field magnitude created by the sources at the flat (output) edge.

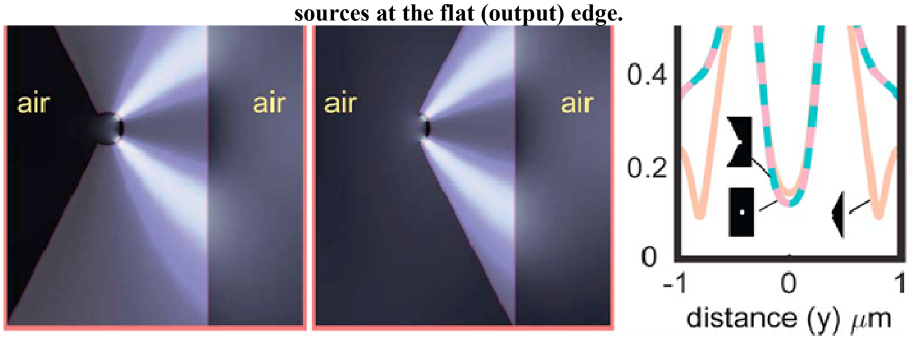


Figure 5: Designs for open-body hyperlenses. (a) Magnetic field map inside a ¾ -body lens. (b) Field map inside a ¼ -body lens. Shaded areas indicate the lens cross-section in (a) and (b). (c) H-field generated by the sources along the flat surface of either the closed or open lens designs.

index material with a Kerr-type nonlinearity and a sub-wavelength layer of linear negative-index material sandwiched between semi-infinite linear dielectrics. We have found that a thin layer of negative-index material leads to significant changes in the hysteresis width when the nonlinear slab is illuminated at an angle near that of total internal reflection. Unidirectional diode-like transmission with an enhanced operational range was demonstrated. These results may be useful for negative-index material characterization and for designing novel devices based on negative-index materials.

From our work and that of other groups, it is evident that optical metamaterials consisting of metal-dielectric composites create complicated systems that are not amenable to analytical solutions. This presents a challenge in optimizing these intricate systems. We have developed applications of three nature-inspired, stochastic optimization techniques in conjunction with fast numerical electromagnetic solvers to design a metamaterial that satisfies predetermined required criteria. In particular, three stochastic optimization tools (genetic algorithm, particle swarm optimization, and simulated annealing) have been used to design a low-loss, optical, negative-index metamaterial [21]. A negative refractive index of around $-0.8+0.2i$ is obtained at a wavelength of 770 nm. The particle swarm optimization algorithm is found to be the most efficient in this case.

Loss is always a significant concern in applications of metamaterials. Since they frequently include plasmonic constituents, metamaterials suffer loss from the metal in their designs. Working towards the compensation of loss in metamaterials, we have studied how to compensate the loss inherent in simpler plasmonic devices [22,23]. The compensation of loss in metal by gain in an interfacing dielectric has been demonstrated in a mixture of aggregated silver nanoparticles and Rhodamine-6G dye. An increase of the quality factor of the surface plasmon (SP) resonance was shown by a six-fold enhancement of the Rayleigh scattering. The compensation of plasmonic losses with gain enables a host of new applications for metallic nanostructures, including low- or no-loss negative-index metamaterials. We have also predicted and experimentally observed a suppression of SP resonance in metallic nanoparticles embedded in dielectric host with absorption.

Section IV. Bibliography

1. H.-K. Yuan, U.K. Chettiar, W. Cai, A.V. Kildishev, A. Boltasseva, V.P. Drachev and V.M. Shalaev, A negative permeability material at red light, Optics Express, Vol.15, No.3, pp.1076-1083 (2007).
2. W. Cai, U.K. Chettiar, H.-K. Yuan, V.C. de Silva, A.V. Kildishev, V.P. Drachev and V.M. Shalaev, Metamagnetics with rainbow colors, Optics Express, Vol.15, No.6, pp.3333-41 (2007).
3. V.M. Shalaev, Optical negative-index metamaterials, Nature Photonics, v.1, 41-48 (2007).
4. U.K. Chettiar, A.V. Kildishev, T.A. Klar, and V.M. Shalaev, Negative index metamaterial combining magnetic resonators with metal films, Optics Express, Vol.14, No.17, 7872-77 (2006).
5. V.M. Shalaev, W. Cai, U.K. Chettiar, H.-K. Yuan, A.K. Sarychev, V.P. Drachev, and A.V. Kildishev, Negative index of refraction in optical metamaterials, Optics Letters, Vol.30, No.24, 3356 (2005).
6. A.V. Kildishev, W. Cai, U. K. Chettiar, H-K. Yuan, A.K. Sarychev, V.P. Drachev and V.M. Shalaev, Negative refractive index in optics of metal–dielectric composites, J. Opt. Soc. Am. B, Vol.23, No.3 423-433 (2006).
7. D.-H. Kwon, D.H.Werner, A.V. Kildishev and V.M. Shalaev, Near-infrared metamaterials with dual-band negative-index characteristics, Optics Express, Vol.15, No.4, pp.1647-52 (2007).
8. V.P. Drachev, W. Cai, U. Chettiar, H.-K. Yuan, A.K. Sarychev, A.V. Kildishev, G. Klimeck and V.M. Shalaev, Experimental verification of an optical negative-index material, Laser Phys. Lett. 3, No. 1, 49–55 (2006).
9. U.K. Chettiar, A.V. Kildishev, H.-K. Yuan, W. Cai, S. Xiao, V.P. Drachev and V.M. Shalaev, Dual-band negative index metamaterial: double negative at 813nm and single negative at 772nm, Optics Letters, Vol.32, No.12, pp.1671-73 (2007).
10. D.H. Werner, D.-H. Kwon, I.-C. Khoo, A.V. Kildishev and V.M. Shalaev, Liquid crystal clad near-infrared metamaterials with tunable negative-zero-positive refractive indices, Optics Express, Vol.15, No.6, pp.3342-47 (2007).
11. X. Wang, D.-H. Kwon, D.H. Werner, I.-C. Khoo, A.V. Kildishev and V.M. Shalaev, Tunable optical negative-index metamaterials employing anisotropic liquid crystals, Appl. Phys. Lett., Vol. 91, p. 143122 (2007).

12. A.K. Popov, V.V. Slabko and V.M. Shalaev, Second harmonic generation in left-handed metamaterials, Laser Phys. Lett. 3, No. 6, 293–297 (2006).

13. A.K. Popov and V.M. Shalaev, Negative-index metamaterials: second-harmonic generation, Manley–Rowe relations and parametric amplification, Appl. Phys. B, Vol.84, pp. 131-37 (2006).

14. A.K. Popov, and V.M. Shalaev, Compensating losses in negative-index metamaterials by optical parametric amplification, Optics Letters, Vol.31, No.14, pp. 2169-2171 (2006).

15. W. Cai, U.K. Chettiar, A.V. Kildishev, G.W. Milton and V.M. Shalaev, Nonmagnetic cloak with minimized scattering, Appl. Phys. Lett., Vol.91, p.111105 (2007).

16. V.P. Drachev, U.K. Chettiar, A.V. Kildishev, H.-K. Yuan, W. Cai and V.M. Shalaev, The Ag dielectric function in plasmonic metamaterials, Optics Express, Vol.16, No.2, pp.1186-95 (2008).

17. W. Cai, U.K. Chettiar, A.V. Kildishev and V.M. Shalaev, Designs for optical cloaking with high-order transformations, Optics Express, Vol. 16, No. 8, pp. 5444-52 (2008).

18. A.V. Kildishev, U.K. Chettiar, Z. Jacob, V.M. Shalaev and E.E. Narimanov, Materializing a binary hyperlens design, Appl. Phys. Lett., Vol. 94, p. 071102 (2009).

19. A.V. Kildishev and V.M. Shalaev, Engineering space for light via transformation optics, Opt. Lett. 33, 43 (2008); this paper has been also selected for the January 28, 2008 issue of Virtual Journal of Nanoscale Science & Technology.

20. N. M. Litchinitser, I. R. Gabitov, A. I. Maimistov, V. M. Shalaev, Effect of an optical negative index thin film on optical bistability, Optics Letters, Vol.32, No.2, pp.151-3 (2007).

21. A.V. Kildishev, U.K. Chettiar, Z. Liu, V.M. Shalaev, D.-H. Kwon, Z. Bayraktar and D. H. Werner, Stochastic optimization of low-loss optical negative-index metamaterial, J. Opt. Soc. Am B, Vol. 24, Issue 10, pp. A34-A39 (2007); also selected for publication in the Virtual Journal of Nanoscale Science & Technology (Oct. 22, 2007).

22. M.A. Noginov, G. Zhu, M. Bahoura, J. Adegoke, C. Small, B.A. Ritzo, V.P. Drachev and V.M. Shalaev, The effect of gain and absorption on surface plasmons in metal nanoparticles, Applied Physics B: Lasers and Optics, Vol.86, No.3, pp.455-460 (2006).

23. M. A. Noginov, G. Zhu, M. Bahoura, J. Adegoke, C. E. Small, B. A. Ritzo, V. P. Drachev and V. M. Shalaev, Enhancement of surface plasmons in an Ag aggregate by optical gain in a dielectric medium, Optics Letters, Vol.31, No.20, 3022-24 (2006); see also article highlighting this research; Surface plasmons: Enhanced nanoplasmonics, Nature Photonics, Published online: 5 October 2006.