Negative-Index Materials: Optics by Design

Wounjhang Park and
Jinsang Kim, Guest Editors

Abstract

Index of refraction, a fundamental optical constant that enters in the descriptions of almost all optical phenomena, has long been considered an intrinsic property of a material. However, the recent progress in negative-index material (NIM) research has shown that the utilization of deep-subwavelength-scale features can provide a means to engineer fundamental optical constants such as permittivity, permeability, impedance, and index of refraction. Armed with new nanofabrication techniques, researchers worldwide have developed and demonstrated a variety of NIMs. One implementation uses a combination of electric and magnetic resonators that simultaneously produce negative permittivity and permeability, and consequently negative refractive index. Others involve chirality, anisotropy, or Bragg resonance in periodic structures. NIM research is the beginning of new optical materials research in which the desired optical properties and functionalities are artificially generated. Clearly, creating negative index materials is not the only possibility, and the most recent developments explore new realms of materials with near-zero indexes and inhomogeneous index profiles that can produce novel phenomena such as invisibility. Furthermore, the concept of controlling macroscopic material properties with a composite structure containing subwavelength-scale features extends to the development of acoustic metamaterials. By providing a review of recent progress in NIM research, we hope to share the excitement of the field with the broader materials research community and also to spur new ideas and research directions.

Introduction

Index of refraction, or refractive index, is a fundamental constant describing the interaction between light and material. Index of refraction quantifies, for example, how fast light travels in a material and how strongly a material reflects light on its surface. Vacuum is the reference medium, with unity index of refraction. In a material, electrons and atoms interact with the electromagnetic field of light, giving rise to an index of refraction specific to the material.

Although index of refraction is generally frequency-dependent, all naturally occurring materials are known to have indexes of refraction that are greater than 1. Is it possible to have a negative index of refraction? This question was pondered as early as 1904.[1] No physical principle prohibits negative index of refraction. In 1968, Veselago theorized that a material with negative permittivity and permeability should have a negative index of refraction and that such a material should exhibit a reverse Doppler effect, a reverse Cherenkov effect, and reversed focusing properties in lenses.[2] Negative-index materials (NIMs), however, remained in the realm of purely theoretical imagination until Pendry's seminal article[3] in 2000 ignited major research activities worldwide. In that work, Pendry predicted the possibility of a superlens that could focus light to a very small spot. Moreover, recent advances in nanotechnology have made it possible to design and fabricate artificial structures that can exhibit a negative index of refraction. NIMs are thus now an exciting reality with numerous opportunities for the discovery of new phenomena and the development of novel devices.

Negative Index of Refraction

Index of refraction n is traditionally defined as the ratio of the speed of light in a vacuum c to that in the material ν:

$$n = \frac{c}{\nu}. \tag{1}$$

Constructing the wave equation from Maxwell's equations, one can relate the index of refraction to the relative permittivity ε and relative permeability μ of the material,

$$n^2 = \varepsilon\mu, \tag{2}$$

where the relative permittivity ε and relative permeability μ are defined by the constitutive equations,

$$\mathbf{D} = \varepsilon\varepsilon_0\mathbf{E} \tag{3}$$

and

$$\mathbf{B} = \mu\mu_0\mathbf{H}. \tag{4}$$

In Equations 3 and 4, **D** is the electric displacement field, **E** is the electric field, **B** is the magnetic field, and **H** is the magnetizing field.

Because most naturally occurring materials have values of ε and μ that are equal to or greater than 1, it is generally assumed that the index of refraction is found by taking the positive root of $\varepsilon\mu$; that is,

$$n = \sqrt{\varepsilon\mu}. \tag{5}$$

An important exception to this general rule is metals. Because of the large density of free electrons, a metal exhibits negative ε and positive μ in the visible and lower-frequency ranges of the electromagnetic spectrum when the frequency is below the metal's characteristic plasma frequency. This results in an imaginary value of n, leading to exponentially decaying waves instead of propagating waves. An exponentially decaying wave is called an evanescent wave, and this explains why most metals are highly reflecting, rather than transparent.

In 1968, Veselago considered the possibility of a material with simultaneously negative values of ε and μ.[2] Negative permittivity requires electric resonance in the material, and negative permeability requires strong magnetic resonance. When both ε and μ are negative, the index of refraction is real, so that the material should support propagating waves. Consider a plane wave in such a

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material. Maxwell's curl equations can be written as

$$\mathbf{k} \times \mathbf{E} = \omega\mu_0\mu\mathbf{H} \tag{6}$$

and

$$\mathbf{k} \times \mathbf{H} = -\omega\varepsilon_0\varepsilon\mathbf{E}. \tag{7}$$

In a positive-index material with positive ε and μ, the wave vector **k** and the electric and magnetic field vectors **E** and **H** form a right-handed set. However, in a negative-index material where ε and μ are both negative, the **k**, **E**, and **H** vectors form a left-handed set. In that case, the wave vector **k**, which represents the direction of propagation of phase fronts and the Poynting vector,

$$\mathbf{S} = \mathbf{E} \times \mathbf{H}, \tag{8}$$

which represents the direction of energy flow, are antiparallel. Thus, if we choose the direction of energy flow as the reference propagation direction, the index of refraction must be negative so that the wave vector **k** is in the opposite direction to the energy flow.

At this point, it is useful to make a distinction between the velocities of the energy flow and the phase front. The rate of energy flow is related to the group velocity ν_g of the electromagnetic wave. This is the velocity at which the envelope propagates when a combination of plane waves forms a wave packet. On the other hand, the magnitude of wave vector **k** is given by the phase velocity ν_p as

$$k = \omega/\nu_p = n\omega/c, \tag{9}$$

where ω is the angular frequency. The phase velocity, which is the velocity used in Equation 1, represents the speed at which phase fronts move. Thus, in a negative-index material, the phase velocity and group velocity have opposite signs, and the phase fronts move backward compared to the direction of energy flow. Figure 1 illustrates the propagation of a wave packet in a NIM, where the packet envelope and phase fronts propagate in opposite directions.

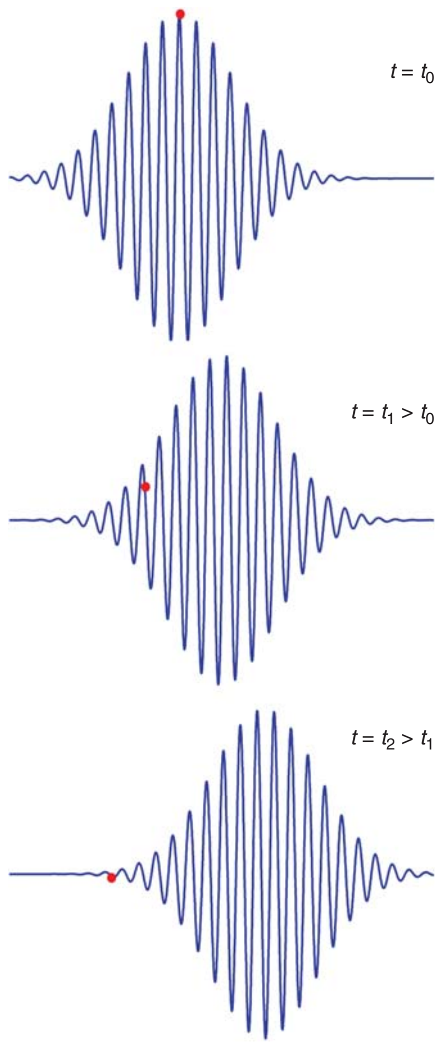

Figure 1. Propagation of a Gaussian wave packet in a negative-index material as time proceeds from t_0 through t_1 to t_2. The envelope of the packet propagates to the right, whereas the phase fronts move to the left. To guide the readers' eyes, a constant phase point in each snapshot is marked by a red dot.

Superlenses

In 2000, Pendry reported that a slab of NIM can focus light using both propagating and evanescent waves, leading to spatial resolutions well below the wavelength.[3] In regular optics, an image is constructed by capturing only the propagating waves. Because the evanescent waves decay exponentially, any information they carry is lost. A simple Fourier analysis shows that the evanescent waves carry information on a length scale smaller than the light wavelength. Thus, in conventional optics, the reconstructed image is missing any details smaller than the wavelength. This is called the diffraction limit and is generally considered the fundamental limit on the achievable spatial resolution in an optical imaging device.

Pendry noted that, in a slab of NIM, the exponentially decaying evanescent wave becomes an exponentially increasing wave. The physical origin of enhanced evanescent wave transmission is coupling with the surface mode at the interface between a NIM and a positive-index material. As a result, a substantial amplitude of evanescent waves exists at the image position, contributing to the reconstruction of the image and providing subwavelength-scale information (Figure 2). This enables the NIM lens to achieve super-resolution, for which reason it is often called a "superlens."

In the extreme near-field limit, also referred to as the quasistatic limit, where all length scales involved in the imaging system are much smaller than the wavelength, the equations for reflection and transmission become independent of permittivity for s-(or transverse electric) polarization and of permeability for p-(or transverse magnetic) polarization. It therefore follows that, for p-polarized light, a slab of material with negative permittivity and positive permeability will act as a superlens because of the permittivity. On the basis of this principle, super-resolution was demonstrated using a thin silver film for ultraviolet light[4,5] and a silicon carbide film for infrared light.[6]

Although the prospect of achieving super-resolution without a scanning mechanism is exciting, the operation of a superlens is generally limited to the near-field region: both the object and image must be in close proximity to the lens itself. Near-field operation can still enable novel applications such as contact lithography. However, achieving far-field operation would greatly expand the utility of the superlens. The near-field restriction is fundamentally related to the nature of evanescent waves, which decay exponentially outside the superlens and thus cannot carry high-resolution information much farther than the wavelength. Therefore, far-field operation requires some mechanisms that convert the evanescent waves to propagating waves without losing the high-resolution information.

One mechanism that can accomplish this is to use a grating coupler.[7] A properly designed grating can translate a range of evanescent waves into the propagating regime, enabling them to propagate into the far-field region. Once detected in the far-field region, the grating-translated evanescent waves can be Fourier transformed back to the original Fourier components so that the image can be properly reconstructed.

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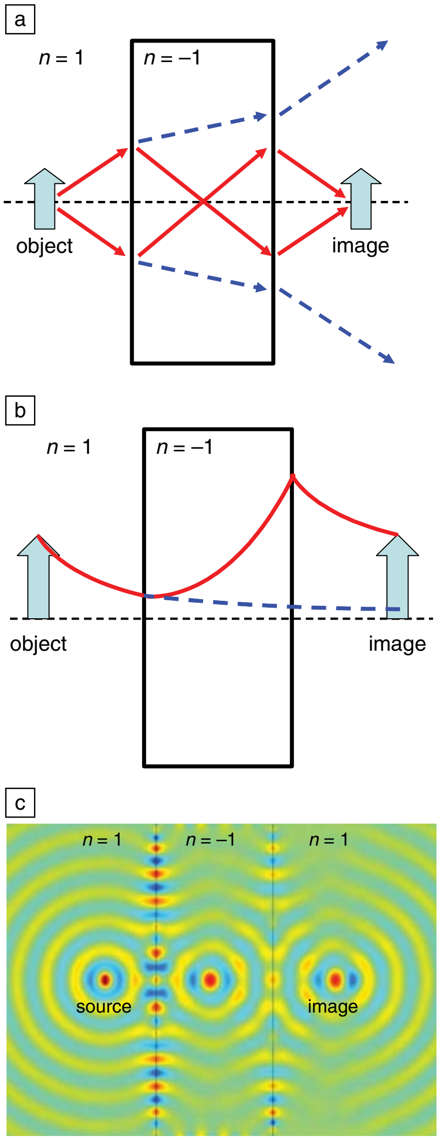

Figure 2. (a) Ray diagram (red arrows) showing the propagating waves being brought to a focus because of negative refraction at the interfaces. Ray paths for a positive-index slab are shown by blue arrows for comparison. (b) Amplitude of evanescent wave (red solid line) illustrating enhanced transmission of evanescent waves. The case for a positive-index slab is shown by the blue dashed line for comparison. (c) Full wave simulation of point-source imaging by a negative-index lens. The wavefronts clearly show the internal and external focus formed by the negative refraction experienced by the propagating waves. The fact that the image size is much smaller than the wavelength indicates enhanced evanescent wave transmission.

Alternatively, one can use a multilayer of superlenses in a cylindrical geometry.[8] In this case, the evanescent waves at the inner surface of the cylindrical lens are converted into propagating waves by the time they reach the outer surface. Such a lens also provides magnification according to the ratio of inner and outer radii. This type of lens is called a "hyperlens" because the effective medium description of the multilayer structure exhibits a hyperbolic dispersion surface. A hyperlens composed of a multilayer of silver and Al_2O_3 was recently demonstrated.[9]

Negative-Index Materials

According to Veselago's prescription, a negative index requires the permittivity and permeability to simultaneously be negative. Negative permittivity is relatively easy to find in natural materials such as metals at frequencies below their plasma frequencies and materials with strong phonon resonance near their phonon frequencies. Negative permeability, which requires a strong magnetic resonance in the material, is much more difficult, especially at frequencies greater than the gigahertz range, and thus requires artificial structures that produce a magnetic response at high frequencies. Pendry et al. proposed split-ring and swiss-roll structures whose internal capacitance and inductance produced a significant magnetic response.[10] Split rings, combined with a metal wire array that provides negative permittivity, were used in the first experimental demonstration of negative refraction in the microwave frequency region[11] and quickly became the most widely used structure for magnetic resonance.

The most common design of a split-ring resonator consists of two concentric metal rings, typically copper, for example. Each ring has a split, and the splits are placed on opposite sides. The separation between the inner and outer rings is much smaller than their radii. The magnetic field of incident light induces current around the rings. However, because of the split, current cannot flow around a single ring but is capacitively coupled to the other ring. Consequently, this system exhibits a characteristic resonant frequency determined by its inductance and capacitance, leading to a resonant behavior in effective permeability.

A variety of split-ring designs have been investigated, and analytical and numerical models have been developed. It should be noted that the split-ring medium generally exhibits large magnetoelectric coupling.[12] Consequently, the split-ring structures support electric (magnetic) resonances excited by a magnetic (electric) field, and this leads to complex cross-polarization effects. To avoid such effects, a modified split-ring resonator consisting of two identical split rings that have splits on opposite sides and are separated by a thin dielectric layer was proposed.[13]

The resonance frequency of a split-ring resonator is inversely proportional to the linear dimension of the split rings. Therefore, reducing the size of the split rings is a straightforward way to increase the resonance frequency. Using this approach, the magnetic activity induced by split-ring structures has been demonstrated for terahertz,[14] mid-infrared,[15] and near-infrared frequency ranges.[16] However, the linear scaling fails in the visible frequency range, and most metals exhibit increased loss in the visible range, further weakening the magnetic resonance. New designs are therefore needed for operation at visible frequencies.

The most successful design has been a pair of metal nanorods that can support an antiparallel plasmon resonance and thus produce a magnetic moment.[17] A refractive index of –0.3 was soon demonstrated in an array of gold nanorod pairs fabricated by electron beam lithography.[18] The combination of nanorod pairs with long metal wires has also been proposed to produce a negative-index material.[19] In this scheme, a magnetic response is produced by the nanorods pairs, and an electrical response is produced by the metal wires. This structure can also be viewed as a pair of voids in a metal background. The optical properties are expected to be similar to those of nanorods pairs by the Babibet principle, which states that an aperture and a disk produce an identical diffraction pattern. Using this structure, a negative index was observed in the near-infrared range[18,20] and at 780 nm.[21] Recent progress in this area is reviewed by Chettiar et al. in this issue.

The approaches described thus far involve two types of resonators, electric and magnetic, to simultaneously produce negative permittivity and permeability. This makes the fabrication very difficult, and manufacturability remains one of the most significant challenges in NIM research. Alternatively, it is possible to construct NIMs with only one type of resonator. For example, negative refraction can occur when electric resonators are

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embedded in a chiral medium, where two orthogonal circular polarizations experience different values of refractive index. When electric resonators are embedded in a chiral material, the chirality shifts the dispersion curves and produces a frequency region in which the sign of the group velocity is opposite to that of the phase velocity, which is a characteristic of negative refraction.[22]

It is also possible to incorporate strongly anisotropic material to induce negative refraction. In an anisotropic material, the permittivity is different for different propagation directions. This results in nonparallel **E** and **D** vectors, which subsequently causes the Poynting vector **S** (Equation 8) to deviate from the wave vector **k**. When the vector difference between **S** and **k** is large enough, it is possible for **S** to point in the opposite direction to **k**, which is the signature of negative refraction. Recently, a semiconductor multilayer stack was designed to exhibit negative permittivity along the optical axis and positive permittivity perpendicular to the optical axis, thus providing an experimental demonstration of negative refraction.[23]

Yet another mechanism to achieve negative refraction is to use the Bragg resonance in a photonic crystal, which refers to a material with a periodic refractive index profile. The multiple reflections due to the periodicity strongly modulate the light propagation and can produce many novel optical properties such as photonic bandgaps, superprisms, self-collimation, and negative refraction.

Photonic crystals can exhibit negative refraction through two distinct mechanisms. In the first case, the photonic crystal has a positive effective refractive index but exhibits negative refraction because of the negative curvature of the dispersion surface.[24] In this case, **S** and **k** exhibit a large vector difference from each other, and the photonic crystal behaves the same way as in the strongly anisotropic material described previously.

The second possibility is that the photonic crystal actually exhibits an isotropic dispersion surface with a negative gradient.[25] Superlensing is possible in both cases. Negative-index imaging by a negative-index photonic crystal was demonstrated first in the microwave region[26] and then more recently in the near-infrared region.[27] Furthermore, the reversal of phase fronts due to the negative curvature of the dispersion surface was directly measured by interferometric near-field scanning optical microscopy.[28] Silicon-based photonic crystal structures that can be fabricated and integrated with other silicon-based optical and electronic devices are a promising route for negative-index materials. The recent progress in this area is reviewed by Baba et al. in this issue.

The greatest challenge in NIM research, particularly for optical applications, is in fabrication. In most implementations, NIMs require deep-subwavelength-scale features, which are in the nanometer range for optical-frequency operation. So far, all experimental demonstrations of optical-frequency NIMs have involved elaborate nanoscale fabrication by electron beam lithography, which is difficult to envision being scaled up for large-scale manufacturing. It also limits the achievable structures to planar two-dimensional structures and is not suitable for three-dimensional bulk NIMs. There have been exciting new developments involving nanoimprinting and stamping that greatly enhance the manufacturability of NIMs. The recent progress in this area is reviewed in this issue by Chaturvedi et al.

A New Frontier in Materials Science

Materials science is important to research in NIMs for many reasons. Fabrication of a NIM requires precise synthesis, patterning, and/or directed assembly of nanoscale materials, each of which critically relies on progress in materials research. Studies on bottom-up fabrication approaches for NIMs are scarce, and we believe that fabrication based on self-assembly, for example, would be highly beneficial. More importantly, NIMs represent a new class of composite materials in which the macroscopic properties are engineered by the constituent materials and structures. In fact, the latest trend in NIM research goes beyond achieving negative indexes of refraction to producing high indexes,[29] near-zero indexes,[30] and inhomogeneous index profiles in order to achieve invisibility,[31] opening an entirely new field called transformation optics.

The new concept of by-design composite materials, often referred to as metamaterials, provides an unprecedented array of opportunities in the design and development of new functional materials and devices. It blurs the traditional distinction between materials and devices: Materials are no longer mere ingredients used to make devices; rather, materials themselves are now sophisticated enough to deliver desired functionalities and interface with other materials. Although current research activities on metamaterials are heavily focused in optics and photonics, the concept can be extended to other functional materials. An excellent example is an acoustic metamaterial constructed of mechanical resonators.[32] This structure exhibits a negative bulk modulus by driving mechanical oscillators at frequencies slightly above the resonance frequency. By combining this negative bulk modulus with a negative effective mass density, negative refraction for acoustic waves can be achieved. Fok et al. review this important emerging area in this issue.

We thus see NIM research as an important new frontier in materials research. In this issue of *MRS Bulletin*, we describe the latest developments in NIM research in hopes that they will inspire materials scientists in many areas of specialization and spur new research activities in broader fields of materials science.

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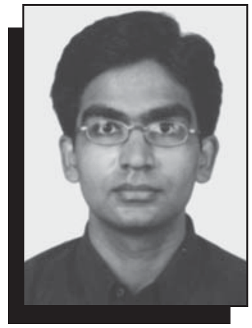

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