In 2000, an artificially structured composite was shown to exhibit a negative index of refraction over a band of microwave frequencies. This demonstration of a previously unexploited electromagnetic (EM) property proved the importance of engineered materials and has resulted in an extraordinary amount of continued development by scientists working in optics, electromagnetism, physics, engineering, and materials science. Experimental results revealing new phenomena and potential applications for these artificial materials, more recently termed metamaterials (MMs), have provided the basis for impressive growth in this burgeoning field. EM-MMs exhibit exotic properties not easily achieved using naturally occurring materials. In addition to a negative index (NI) of refraction, properties such as artificial magnetism, negative permittivity, and negative permeability have been observed in fabricated MM composites; these material properties are either absent from conventional materials or are difficult to achieve over various bands of the EM spectrum. With encouraging results having been demonstrated at microwave frequencies, there has been a determined push to extend MMs to the terahertz, infrared, and visible bands. The pursuit of these higher frequency MMs has led to the establishment of many research programs, conferences, workshops, and significant funding efforts dedicated to understanding and developing MMs. Progress has been rapid: although the entire subject of MMs is relatively new, the scaling of artificial structures has already been demonstrated from radio frequencies (RFs) to millimeter-wave frequencies.

Willie J. Padilla†*, Dimitri N. Basov#, David R. Smith§
†Materials Science & Technologies-Center for Integrated Nanotechnologies, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
#Physics Department, University of California San Diego, San Diego, CA 92037, USA
§Department of Electrical & Computer Engineering, Duke University, Durham, NC 27708, USA
*E-mail: willie@lanl.gov

ISSN: 1369-7021 © Elsevier Ltd 2006

Negative refractive index metamaterials
far infrared (FIR), mid-infrared (MIR), and near infrared (NIR) wavelengths, spanning nearly seven orders of magnitude in frequency. With MM design and fabrication procedures becoming increasingly routine, the prospect of designer materials with a range of new and selectable EM properties at nearly any wavelength band is nearing reality. By providing access to new realms of material response, MMMs have and will continue to impact the fields of physics, materials science, engineering, optics, nanotechnology, and many others branches of science. Since 2000, the rate of publications on EM-MMs has grown exponentially, indicative of the intense interest that artificial materials have generated. Much of that interest has been sparked by the prospect of MMMs with negative refractive index — a property not found in nature. In this review, we focus on artificial structures formed by repeated elements whose physical dimension and spacing are less than the wavelengths of interest, and can be treated as homogeneous to good approximation. For more detailed information on MMMs and NI, including the prospects of achieving negative refraction in photonic crystals and other systems, the reader is referred to several review articles12-14 and books15-17 on the subject.

EM response of materials

To understand MMMs, it is necessary to understand material response to EM waves in general. EM response in homogeneous materials is predominantly governed by two parameters. One of these parameters, $\varepsilon(\omega)$, describes the response of a material to the electric component of light (or other EM wave) and the other, $\mu(\omega)$, to the magnetic component at a frequency $\omega$. Both of these parameters are typically frequency-dependent complex quantities, and thus there are in total four numbers that completely describe the response of an isotropic material to EM radiation at a given frequency,

$$
\varepsilon(\omega) = \varepsilon_r(\omega) + i\varepsilon_i(\omega)
$$

$$
\mu(\omega) = \mu_r(\omega) + i\mu_i(\omega)
$$

For most materials, the two complex quantities $\varepsilon$ and $\mu$ are the only relevant terms and hence dictate the response between light and matter. Among the various fields of science, however, there are many other EM parameters used to describe wave propagation that are related to the material parameters shown in eq 1 by simple algebraic relations, for example such quantities as the absorption or the conductance of a material can be redefined in terms of $\varepsilon$ and $\mu$.

A commonly used EM parameter is that of the index of refraction, which is defined as $n(\omega)^2 = \varepsilon(\omega)/\mu(\omega)$. The index of refraction provides a measure of the speed of an EM wave as it propagates within a material. In addition, the refractive index also provides a measure of the deflection of a beam of light as it crosses the interface between two materials having different values for their refractive indices. The quantitative measure of this bending was provided by Willebrord Snell in 162118,19, who showed that,

$$
n_1 \sin \theta_1 = n_2 \sin \theta_2
$$

where $\theta_1$ and $\theta_2$ are the angles the light ray makes with the surface normal of each media. A simple ray tracing diagram is shown in Fig. 1 for rays emanating from a point source in free space, incident on a slab with positive index of refraction.

The index of refraction of the first and second media is denoted by $n_1$ and $n_2$ respectively, and $\theta_1$ and $\theta_2$ are the angles the light ray makes with the surface normal of each media. A simple ray tracing diagram is shown in Fig. 1 for rays emanating from a point source in free space, incident on a slab with positive index of refraction. On reaching the surface of the slab, the rays emanating from the source bend at the interface between free space and the glass with an angle as determined by Snell’s Law (eq 2).

$$
n_1 \sin \theta_1 = n_2 \sin \theta_2
$$

In virtually all undergraduate and graduate level texts on the subject of optics or electricity and magnetism the refractive index is always assumed positive. But nature has hidden a great secret from us, first described by Russian physicist Victor Veselago. Veselago realized that if a material were found that had negative values for both the electric and magnetic response functions, i.e. $\varepsilon(\omega)<0$ and $\mu(\omega)<0$, then its index of refraction would also be negative, $n(\omega)<0$.

Although Veselago conjectured that naturally occurring materials with negative refractive index might be found or synthesized in naturally occurring materials, such materials have never been found. However, because artificially structured MMMs can have controlled magnetic and electric responses over a broad frequency range, it is possible to achieve the condition $\varepsilon<0$ and $\mu<0$ in artificial composites and Veselago’s hypothesized material can, indeed, be realized.

**Electromagnetic metamaterials**

So what are these fantastic artificial materials capable of achieving such a ‘rare’ state of nature? Next we overview these increasingly common materials and explain how they are used to achieve unique response. The term metamaterial (MM) refers to an artificially
constructed material or composite having distinct and possibly superior properties as compared with the constituent materials from which it is composed. Other types of media exist to which this term might equally well apply. Photonic crystals, for example, are periodic dielectric or metallic structures capable of achieving negative phase velocity and thus NI. However, these structures are not easily described by bulk parameters such as $\varepsilon$ and $\mu$, and hence we exclude them from our discussion.\textsuperscript{20-23} Rather, we are concerned here with those artificial structures that can be viewed as homogeneous, described by values of $\varepsilon$ and $\mu$. The desired material consists of an array of subwavelength elements, designed independently to respond preferentially to the electric or magnetic component of an EM wave. To describe the conceptual basis of a NI MM, it is first useful to summarize the design of the constituent magnetic and electric elements that respectively give rise to negative $\mu$ and negative $\varepsilon$.

**Magnetic response**

The split ring resonator (SRR) has been the element typically used for response to the magnetic component of the EM field. This ‘magnetic atom’ was proposed by Pendry in 1999.\textsuperscript{19} In Fig. 2a we show a schematic of this MM and in Fig. 2b how this element is arranged in an array to form an effective magnetic material. In the simplest representation, the SRR can be thought of as an LC resonator. A time varying magnetic field polarized perpendicular to the plane of the SRR will induce circulating currents according to Faraday’s law. Because of the split gap in the SRR, this circulating current will result in a build up of charge across the gap with the energy stored as a capacitance. The SRR can thus be viewed as a simple LC circuit, with a resonance frequency of $\omega_0^2/1/LC$, where the inductance results from the current path of the SRR. For frequencies below $\omega_0$, currents in the SRR can keep up with the driving force produced by the externally varying magnetic field and a positive response is achieved. However, as the rate of change (frequency) of the external magnetic field is increased, the currents can no longer keep up and eventually begin to lag, resulting in an out-of-phase or negative response.

The general form of the frequency dependent permeability of the SRR\textsuperscript{19} has the generic form

$$\mu_{eff}(\omega) = 1 + \frac{F}{\omega^2} \left( \frac{\omega_0^2 - \omega^2 - i\Gamma \omega}{\omega^2} \right)$$

Fig. 2 Elements used for construction of MMs. In (a) we show an SRR with an external magnetic field incident upon it. When the SRR shown in (a) is arranged into an array (b), it behaves as an effective material and described by a magnetic response. In (c) we show a medium used for electric response, the straight wire medium. A new element used for electric response is shown in (d). The orientation of the external electric field is shown. This new electric particle is also arranged in a planar array for an effective response.
Negative refractive index metamaterials

REVIEW FEATURE

Box 1

The index of refraction is a product of two complex functions, $\epsilon(\omega)$ and $\mu(\omega)$. By representing the magnetic and electric response functions by Lorentz oscillators (eq 4) in complex form we see that the index squared is $n^2 = \epsilon(\omega)\mu(\omega) = \epsilon_0 \mu_0 n_0^2 1 - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma}$. The complex index of refraction then becomes $n = \sqrt{\epsilon(\omega)\mu(\omega)}$. Note the phase of the index of refraction is simply the average of the phases of the magnetic permeability and the electric permittivity, i.e. $\theta_n = (\theta_\epsilon + \theta_\mu)/2$. This indicates that the vector describing the index of refraction must lie in quadrant 3 of the complex space. Thus finally we see that although there is ambiguity in which sign to take for the real part of the index, i.e. $n = n_1 + in_2 = \pm\sqrt{\epsilon(\omega)\mu(\omega)}$, when we consider causal functions it is clear that the index of refraction is required to be negative $n_1 < 0$.

where $F$ is a geometrical factor, $\omega_0$ is the resonance frequency, and $\Gamma$ is the resistive damping factor. Note that this magnetic response function has real and imaginary frequency dependent parts. In Box 1, the frequency dependent permeability of the SRR medium for typical parameters is plotted, showing the frequency dependent resonant form. If the ‘strength’ of the resonance is great enough and the damping small enough, the SRR can yield a negative magnetic response. The solid blue curve in Box 1 corresponds to the real part of $\mu$, which displays a region of negative values for this example.

Electric response

Naturally occurring materials that yield a negative response to the electric component of light have been known for several decades. Any metal below its plasma frequency (the frequency at which it becomes transparent) yields negative values of the permittivity. This $\epsilon_1 < 0$ response results from the free electrons in the metal that screen external EM radiation. But a bulk metal is not the only material that exhibits negative electric response; a distributed array of conductors, or even a grating on a conductor, can give the same result. Many decades ago researchers fabricated structures having $\epsilon < 0$ using arrays of conducting wires and other unique shapes. This technology was recently reintroduced with a more physics-oriented understanding. Currently, variations of the wire lattice being used to create $\epsilon < 0$ media include straight wires, cut-wire segments, and loop wires. A straight wire medium is depicted in Fig. 2c. In addition, there have been further advances in the development of electric MMs with new designs analogous to the SRR being demonstrated (Fig. 2d).

The generic frequency dependent permittivity has the form $\epsilon(\omega) = 1 - \frac{\omega_0^2}{\omega^2 + i\omega\Gamma}$ (4) where the plasma frequency, $\omega_0^2$, is $\omega_0^2 = 4\pi\sigma/m^* n$ (5) and $n$ is the carrier density, $\sigma$ is the charge of an electron, and $m^*$ is the effective mass of the charge carriers (usually electrons) and $m^*$ to their effective mass. In a wire MM, $n$ and $m^*$ are related to the geometry of the lattice rather than the fundamental charge carriers, giving MMs much greater flexibility than conventional materials. Because the effective density can be reduced substantially by making the wires thin, which has the added effect of increasing the effective mass of the charge carriers, the effective plasma frequency can be reduced by many orders of magnitude. In the context of NI MMs, the wire lattice and its variants are a convenient means of achieving a medium for which $\epsilon_1 < 0$. Because the plasma frequency can be tuned by geometry, the region of moderately negative values of $\epsilon$ can be made to occur at nearly any frequency range, from low RF to the optical.

Negative index metamaterials

Having identified artificial structures that can separately provide $\epsilon_1 < 0$ and $\mu_1 < 0$, we can combine the two, according to Veselago’s prescription, and construct a material with $n < 0$. But what, if anything, is actually unusual about a NI material?

Veselago pointed out that a medium having an NI of refraction would essentially add a new twist to virtually every EM phenomenon. The phase velocity of a wave is reversed in NI materials; the Doppler shift of a source relative to a receiver is reversed; Cerenkov radiation emitted by a moving charged particle is in the backward rather than the forward direction; radiation pressure is reversed to become a radiation tension; converging lenses become diverging lenses and vice versa. These are just some of the changes to basic EM phenomena that would result in a NI material.

As intriguing as Veselago’s predictions were, naturally occurring materials with a NI were not known at the time and his results remained largely overlooked. However, in 2000 Smith et al. fabricated an NI material using artificially constructed MMs. This NI MM...
Negative refractive index metamaterials

...exhibiting resolution beyond the diffraction limit. However, Pendry showed that a flat slab of NI material could produce a focus with resolution exceeding the diffraction limit. This was predicted by experimental results that were in favor of negative refraction. The first demonstration and confirmation of negative refraction was performed in 2001 by Shelby et al. Negative refraction was determined by a Snell’s Law experiment using a prism shaped MM wedge, as shown in Fig. 3. A beam of microwave radiation incident on the prism was observed to refract to the opposite side of the surface normal, thus demonstrating negative refraction. For reference, the same beam deflection experiment was performed using a Teflon prism (positive index). The positive index sample deflected the beam to the opposite side of the surface normal at an angle consistent with the known index of the material. These initial results have now been confirmed by numerous researchers, including Parazzoli et al. from Boeing and Houck et al. from Massachusetts Institute of Technology. With Veselago’s NI material finally a reality, and with numerous experimental confirmations having established the validity of the MM approach, researchers have taken up Veselago’s exploration of negative refraction. An ever widening array of altered or new phenomena associated with negative refraction are being discovered including, e.g. the reversal of the Goos-Hanchen effect and enhanced diffraction.

Perhaps one of the most striking predictions for MMs came in 2000, when Pendry showed that a flat slab of NI material could produce a focus with resolution exceeding the diffraction limit. This was an extraordinary prediction, since it required that the normally exponentially decaying evanescent terms produced by a source would actually be recovered in the image formed by the slab. All sources of EM radiation possess both propagating components and components that stay fixed, decaying rapidly away from the source. Mathematically, all EM sources can be expressed as a superposition of propagating plane waves and exponentially decaying near-fields. These exponentially decaying terms cannot be recovered by any known positive index lens. Since the near field is responsible for conveying the finest details of an object, their absence limits the resolution of positive index optics to roughly $\lambda/2$ – the diffraction limit. However, Pendry predicted that an NI lens would actually be able to recover the exponentially decaying near-field components at the image, thereby exhibiting resolution beyond the diffraction limit.

In Fig. 4, we summarize some work extending MM response from the initial discovery range – microwave (second row) – to both higher and lower frequency ranges. This microwave experiment was performed near 1 GHz and showed the ability of a planar left-handed lens, with a relative refractive index of -1, to form images that were subwavelength focusing with a NI material. This microwave experiment was performed in 2001 by Shelby et al. Negative refraction was the matter finally settled.

In the right panel of Fig. 1, we show how this focusing occurs for a medium of $n = -1$. In the top portion of Fig. 1, a ray tracing diagram shows how rays are focused by the slab. But the ray tracing picture leaves out the evanescent, or exponentially decaying, components. The diagram in the bottom right of Fig. 1 shows an evanescent component that is, in some sense amplified, by the slab – growing exponentially as a function of distance and then decaying exponentially until it reaches its original magnitude at the image.

In 2004, Grbic and Eleftheriades demonstrated experimentally subwavelength focusing with a NI material. This microwave experiment was performed near 1 GHz and showed the ability of a planar left-handed lens, with a relative refractive index of -1, to form images that overcame the diffraction limit. The NI lens consists of a planar slab constructed from a grid of printed metallic strips over a ground plane, loaded with series capacitors and shunt inductors. The measured half-power beamwidth of the point source image formed by the NI lens is 0.21 effective wavelengths, which is significantly narrower than that of the diffraction-limited image corresponding to 0.36 wavelengths.

Natural materials

While the most familiar examples of NI materials have made use of artificially patterned MMMs, combinations of naturally occurring...
Fig. 4 Summary of MM results from RF to near optical frequencies. In the left column we detail the frequency range in which each MM was demonstrated and note the reference number in this review. The middle column shows a photo of the MM from each publication, and the third column shows some data detailing the MM response. The top row is an investigation of ‘swiss-roll’-type magnetic structures to guide magnetic flux in magnetic resonance imaging machines. The second row is the original work in which NI materials were discovered at microwave frequencies. The third row shows some recent work on MMs at millimeter-wave frequencies. The next column details the first work extending MMs out of the microwave into the terahertz regime. The bottom two columns show further extension of the SRR magnetic MM medium to MIR and NIR frequencies. [Part (a) reproduced with permission from 5. © 2001 American Association for the Advancement of Science (AAAS). Part (b) reproduced with permission from 1. © 2000 American Physical Society (APS). Part (c) reproduced with permission from 6. © 2004 AAAS. Part (d) reproduced with permission from 7. © 2004 AAAS. Part (e) reproduced with permission from 8. © 2004 AAAS. Part (f) reproduced with permission from 10. © 2005 APS.]
materials may yet play a role in negative refraction. Recently, it has been demonstrated that an NI can be exhibited by magnetodielectric spherical particles \(^4^7\), superlattices of natural materials \(^4^8\), and uniaxial crystals \(^4^9\). There are many theoretical suggestions of various other methods one might use to achieve an NI at NIR and optical frequencies \(^5^0,5^1\). It is worthwhile to note that although the NI in these materials comes about ‘naturally’, since these materials are engineered or special cases they can also be considered MMs, as they are constructed in particular shapes and/or combinations. In Fig. 5, we show one example of a NI material constructed from natural elements – insulating magnetodielectric spherical particles embedded in a background dielectric material. The effective permeability and permittivity of the mixture has been shown to be simultaneously negative at a particular frequency, thus exhibiting NI.

**Tunable metamaterials**

As emphasized above, all current implementations of NI media have been accomplished over a narrow frequency range in the vicinity of the resonant frequency \(\omega_0\). The latter parameter is rigidly determined by the geometrical dimensions of the SRR and possibly other elements used to construct NIMs. Both from the viewpoint of applications, as well as for the purpose of the understanding of the intrinsic properties of negative media, it is desirable to implement structures with tunable and reconfigurable resonant properties. Some possible solutions to this intriguing problem have been proposed recently \(^5^2\).

The SRR structure is interesting not just for its magnetic properties, but also because large electric fields can potentially build up in the gap region between the rings. Methods that alter the local dielectric environment, then, have the potential to shift the resonant frequency of the SRR, which has the approximate analytic form \(^5^3\),

\[
\omega_0 = \sqrt{\frac{3}{\pi \mu_0 C r^6}} \tag{6}
\]

where \(r\) is the ring radius, \(\mu_0\) is the static magnetic permeability, and \(C\) is the capacitance per unit area between the two rings. Detailed simulations have confirmed that the resonant frequency of the SRR is indeed very sensitive to the SRR capacitance, which in turn depends on the value of the dielectric constant of the substrate \(\varepsilon_s\). The frequency of operation of the SRR thus scales as \(\sim (1/\sqrt{\varepsilon_s})\) so that \(\omega_0\) can be made tunable by either changing the substrate dielectric or the resonant permeability will shift accordingly.

This form of dynamic tuning has been accomplished recently by Padilla et al. \(^5^3\), who control and modify the substrate dielectric by photodoping an SRR array patterned on an insulating GaAs substrate.

A 50 fs pulse of 800 nm light is used to excite photocarriers across the band gap of the GaAs substrate. Because photodoped charges are relatively long lived (1 ns), the quasi-steady state response of the composite MM sample can be studied using terahertz time domain spectroscopy. Representative data from this work are displayed in Fig. 6. In this particular experiment, the conductivity arising from mobile photocharges shunts the low-frequency resonance at \(\omega_0 = 0.5\ \text{THz}\) associated with circulating (magnetic) currents, whereas the higher energy (electric) mode at \(\omega_1 = 1.6\ \text{THz}\) remains nearly unaffected. This work has revealed the potential of SRR/semiconductor hybrid structures to develop terahertz switches. Response times in the 1-10 ps range would be possible provided materials with faster recombination times are used as the substrate.

Alternative ways of tuning NI materials can be realized by integrating SRR arrays into a metal-insulator-semiconductor (MIS) architecture. Applying a dc electric field between the ring arrays and a semiconducting substrate allows tuning of the dielectric constant of the insulating layer in the MIS device, provided the insulator is fabricated from a high dielectric constant or ferroelectric material \(^5^4\). In order to maintain the electric field across the insulator in the area within the split gaps of the SRRs, it is desirable to fill these structures with semiconducting polymers. Charge injection into a polymer allows one to achieve an electric field in MIS structures over large areas.

---

**Fig. 5** MM constructed from natural materials, magnetodielectric spherical particles embedded in a matrix, which exhibits an NI.

**Fig. 6** Transmission spectra as a function of photodoping fluence for the electric resonance of the SRRs. Free carriers in the substrate short out the gap of the SRR and eventually kill the resonant response \(^5^3\). (Reprinted with permission from \(^5^3\). © 2006 American Physical Society.)
without appreciably changing the dc conductivity of the polymer.\footnote{26} Therefore, in MIS-based devices the dielectric constant and therefore the resonant frequency can be tuned by varying the applied dc voltage. One can envision that each of the rings in an SRR array could be biased with a different voltage. This selective tuning of the dielectric constant could allow one to achieve the controlled distribution of the resonant frequencies over a planar array, thus in principle enabling reconfigurable lenses\footnote{27} and other microwave/terahertz components.

**Outlook: devices and limitations**

In this review we have stressed the novelty of EM-MMs and shown the great flexibility that we now have to design materials with the power to control EM radiation. The ‘knobs’ available to control the two components of EM radiation individually form the basis for such versatility and provide significant advantages over, for example, photonic band gap media. However, there are limitations to the amount of ‘tuning’ of which these materials are capable.

The SMR shown in Fig. 2 depends upon the ‘bulk’ conductivity of a metal. That is, we need macroscopically circulating currents in order to exhibit an effective magnetic response. At optical and ultraviolet wavelengths, metals become transparent to light and thus loose their metallic ‘free-electron’-like properties, including their conductivity. It is expected therefore that EM-MMs will not work at such high frequencies. Also, since wavelength and frequency scale inversely, the cell-size-to-wavelength parameter, $\lambda_0/\lambda < 1$, is no longer satisfied and we are thus not in the effective material regime. Both of the above limitations seem to indicate that EM-MMs will begin to fail for increasing frequencies somewhere around the optical range. However, natural losses in metals will likely contribute to the degradation of the effective material response and the real limitation may be somewhere in the NIR regime.\footnote{28} If we wish to overcome these limitations, we must consider different paradigms in the design of artificial magnetic response or alternatively seek methods to compensate for these losses, i.e. active materials.

**REFERENCES**

6. www.nanotechnology.bilkent.edu.tr/research%20areas/documents/mm-wave.html
10. Ramakrishna, S. A., private communication
17. Hecht, E., Optics, 4th edition, Addison-Wesley, Massachusetts, USA, [2005]