

Negative Refraction at Optical Frequencies in Nonmagnetic Two-Component Molecular Media

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There is significant motivation to develop media with negative refractive indices at optical frequencies, but efforts in this direction are hampered by the weakness of the magnetic response at such frequencies. We show theoretically that a nonmagnetic medium with two atomic or molecular constituents can exhibit a negative refractive index. A negative index is possible even when the real parts of both the permittivity and permeability are positive. This surprising result provides a route to isotropic negative-index media at optical frequencies.

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In 1968 Veselago [1] considered the electrodynamic properties of isotropic media where the real part of the electric permittivity ϵ and the real part of the magnetic permeability μ are simultaneously negative. Veselago showed that if $\epsilon, \mu < 0$ then the electric field \mathbf{E} , the magnetizing field \mathbf{H} and the wave vector \mathbf{k} form a left-handed orthogonal set, contrary to all known naturally occurring materials where the triplet of these vectors is right-handed. Media with both negative electric permittivity and magnetic permeability are referred to as left-handed materials (LHM) [2] at the frequencies for which $\epsilon, \mu < 0$.

A consequence of simultaneously negative ϵ and μ is that the Poynting vector $\mathbf{S} = \mathbf{E} \times \mathbf{H}$ and the wave vector $\mathbf{k} = (\omega/|\mathbf{E}|^2)\mathbf{E} \times \mathbf{B}$ point in opposite directions for a monochromatic plane wave with angular frequency ω , as here the direction of the magnetic field necessarily opposes that of the magnetizing field, $\mathbf{B} = \mu\mathbf{H}$ [1]. Veselago argued that the direction of the energy flow (\mathbf{S}) must point away from its source and thereby reached the surprising conclusion that in LHM the wave vector points toward the source [1]. This in turn led to the prediction that LHM exhibit a negative refractive index, as well as reversed Doppler and Cherenkov effects [1].

No naturally occurring isotropic material is known to have $\epsilon, \mu < 0$ at the same frequency. Pendry *et al.* [3] thus suggested that structures containing metal strips and splitting resonators could be engineered such that both ϵ and μ are negative at microwave frequencies. Shelby *et al.* [4] subsequently reported the observation of negative refraction at 10.5 GHz in a left-handed metamaterial. A major goal of this field of research is the development of media with negative refractive indices at optical frequencies, where exciting concepts such as the “perfect lens” [5] would have their greatest impact. To date, there are no reports of media that exhibit a negative refractive index at optical frequencies. A major impediment to the development of such media is the fact that magnetic resonances occur at much lower frequencies, so magnetic effects at optical frequencies are very weak. Photonic crystals [6,7], and birefringent crystal assemblies [8] have been investi-

gated as dielectric negative-index media, but these are not isotropic and they therefore cannot be characterized by a single, scalar refractive index. In addition, diffraction phenomena may contribute to their properties.

In this Letter we demonstrate that a nonmagnetic system with two molecular components, one of which is in its excited state, can achieve negative refraction. Contrary to common belief, this is possible even when the material is right handed, i.e., the real parts of both ϵ and μ are positive. Such a material could be a route to negative refraction at optical frequencies. The occurrence of negative refraction in isotropic media is especially desirable for refraction experiments and the fabrication of optical components.

There has been much discussion of the directions of energy flux and group velocity in novel optical media. Loudon [9] pointed out that in general these could differ, and as mentioned, Veselago recognized that this could be the case in LHM. Zheleznyakov *et al.* [10] considered this issue in the context of backward electromagnetic waves, and more recently, Pokrovsky and Efros [11] addressed it for LHM. While this discussion contributes much insight to the picture of wave propagation in novel materials, we take a different approach here and focus on the implications of causality for the propagation. We employ a rigorous analysis of causal wave propagation as first discussed by Sommerfeld and Brillouin [12]. We begin by considering the wave equations for a homogeneous isotropic linear medium in a source-free region,

$$\begin{aligned}\nabla^2 \tilde{\mathbf{E}}(\mathbf{r}, \omega) &= -\omega^2 \tilde{\epsilon}(\omega) \tilde{\mu}(\omega) \tilde{\mathbf{E}}(\mathbf{r}, \omega), \\ \nabla^2 \tilde{\mathbf{B}}(\mathbf{r}, \omega) &= -\omega^2 \tilde{\epsilon}(\omega) \tilde{\mu}(\omega) \tilde{\mathbf{B}}(\mathbf{r}, \omega).\end{aligned}\quad (1)$$

$\tilde{\mathbf{E}}(\mathbf{r}, \omega)$ and $\tilde{\mathbf{B}}(\mathbf{r}, \omega)$ are the complex Fourier transforms of the corresponding real fields $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{B}(\mathbf{r}, t)$, where the Fourier transforms in (1) are defined as

$$\tilde{f}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} f(t) e^{i\omega t} dt. \quad (2)$$

The vector equations have a common form for each

Cartesian component and can be simplified to

$$\frac{\partial^2 \tilde{g}(z, \omega)}{\partial z^2} = -\omega^2 \tilde{\epsilon}(\omega) \tilde{\mu}(\omega) \tilde{g}(z, \omega), \quad (3)$$

if we consider a general plane wave propagating in the $+z$ direction. The equation has the time-domain Green's function solutions

$$\begin{aligned} g(z, t) &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \tilde{g}(z, \omega) e^{-i\omega t} d\omega \\ &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{i(\tilde{k}(\omega)z - \omega t)} d\omega. \end{aligned} \quad (4)$$

$\tilde{k}(\omega)$ is in general not single valued due to the branch cuts created in taking the square root, and Einstein causality is needed to determine the correct branch of $\tilde{k}(\omega)$. More precisely, with $\tilde{\epsilon}(\omega) = |\tilde{\epsilon}(\omega)|e^{i\phi_\epsilon}$ and $\tilde{\mu}(\omega) = |\tilde{\mu}(\omega)|e^{i\phi_\mu}$, the two branches are $\tilde{k}(\omega) = \pm \omega \sqrt{|\tilde{\epsilon}(\omega)| |\tilde{\mu}(\omega)|} e^{i(\phi_\epsilon + \phi_\mu)/2}$, with corresponding refractive indices $\tilde{n}(\omega) = \pm c \sqrt{|\tilde{\epsilon}(\omega)| |\tilde{\mu}(\omega)|} e^{i(\phi_\epsilon + \phi_\mu)/2}$. Einstein causality further assures that the physical solution is always

$$\tilde{n}(\omega) = +c \sqrt{|\tilde{\epsilon}(\omega)| |\tilde{\mu}(\omega)|} e^{i(\phi_\epsilon + \phi_\mu)/2}, \quad (5)$$

regardless of the exact functional form of $\tilde{\epsilon}(\omega)$ and $\tilde{\mu}(\omega)$ [13].

We now express the permittivity and the permeability in terms of Lorentz oscillator models as their generality facilitates the discussion of the refractive index for a variety of systems, such as atoms in the gas phase, conductors near a plasmon resonance, or any medium whose optical properties are directly related to the underlying molecular polarizabilities and magnetizabilities. The electric permittivity $\tilde{\epsilon}(\omega)$ and the magnetic permeability $\tilde{\mu}(\omega)$ may then be written as

$$\begin{aligned} \tilde{\epsilon}(\omega) &= \epsilon_0 \left(1 + \frac{F}{\omega_{\text{pole}_\epsilon}^2 - (\omega + i\Gamma)^2} \right) \\ &= \epsilon_0 \frac{(\omega + i\Gamma)^2 - \omega_{\text{zero}_\epsilon}^2}{(\omega + i\Gamma)^2 - \omega_{\text{pole}_\epsilon}^2} \end{aligned} \quad (6)$$

$$\begin{aligned} \tilde{\mu}(\omega) &= \mu_0 \left(1 + \frac{G}{\omega_{\text{pole}_\mu}^2 - (\omega + i\Gamma)^2} \right) \\ &= \mu_0 \frac{(\omega + i\Gamma)^2 - \omega_{\text{zero}_\mu}^2}{(\omega + i\Gamma)^2 - \omega_{\text{pole}_\mu}^2}, \end{aligned} \quad (7)$$

where $\omega_{\text{zero}_\epsilon}^2 = \omega_{\text{pole}_\epsilon}^2 + F$, and similarly $\omega_{\text{zero}_\mu}^2 = \omega_{\text{pole}_\mu}^2 + G$. F , G , and Γ are all taken to be real. A system in the ground state corresponds to $F, G > 0$ and an inverted system has $F, G < 0$. The permittivity of the vacuum is ϵ_0 and its permeability is μ_0 . Γ is the half width at half maximum of the Lorentzian spectrum ($\Gamma > 0$). From Eq. (6) we know that the structures of the zeros and poles

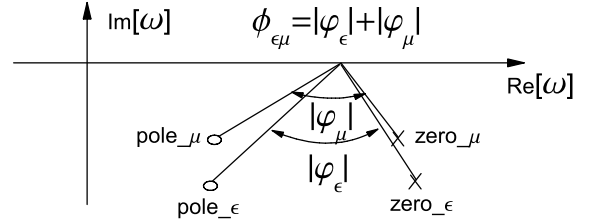


FIG. 1. Zero-pole pairs are shown for a passive LHM ($F > 0$ and $G > 0$) in which both ϕ_ϵ and ϕ_μ have positive signs.

determine ϕ_ϵ , the principle value of the argument of $\tilde{\epsilon}(\omega)$, i.e., $\phi_\epsilon = \text{Arg}(\tilde{\epsilon}(\omega))$. In the case of a noninverted (passive) Lorentz oscillator, the contribution from the zero-pole pair yields a positive ϕ_ϵ , whereas for the inverted system, ϕ_ϵ is negative; and similarly for $\tilde{\mu}(\omega)$. The angles $\phi_{\epsilon\mu} = (\phi_\epsilon + \phi_\mu)$ and $\phi_n = \phi_{\epsilon\mu}/2$ are thus determined by the zeros and poles of both $\tilde{\epsilon}$ and $\tilde{\mu}$. Figure 1 gives an example of how the angle $\phi_{\epsilon\mu}$ is affected by the zero-pole structures of both $\tilde{\epsilon}$ and $\tilde{\mu}$ [22]. We can also see from Fig. 1 that for a passive LHM, i.e., when $\pi/2 < \phi_\epsilon < \pi$ and $\pi/2 < \phi_\mu < \pi$, we have $\pi/2 < \phi_n < \pi$. This results in $\text{Re}[n] < 0$, as has been shown in prior work on passive LHM (c.f. [1,4,5,20,23–26]). Thus, the sign of the index based on causality reproduces prior results [1,5] under appropriate conditions. A systematic treatment of the possible outcomes for the sign of the index appears in Ref. [28]

One major difficulty in obtaining a negative refractive index at optical frequencies through passive LHM has been that for most systems, the magnetic response at such high frequencies is very weak. By recognizing that it is the *combined* contribution from the zero-pole structures of $\tilde{\epsilon}(\omega)$ and $\tilde{\mu}(\omega)$ that determines the sign of the refractive index [as is evident in Eq. (5), where the sign is completely determined by $\phi_\epsilon + \phi_\mu$], one can consider the idea of achieving negative refraction with two closely lying poles and zeros from the electrical response and a nonresonant magnetic response. As we demonstrate below, this is not only theoretically possible, but also seems to be experimentally feasible.

We now show that even when both ϵ and μ are positive, i.e., in a right-handed medium (RHM), the structures of the zeros and poles may in certain cases give rise to a negative refractive index. Surprisingly, this can happen for a non-magnetic system. We consider a two-component system with $\mu = \mu_0$ and with

$$\begin{aligned} \tilde{\epsilon}(\omega) &= \epsilon_0 \left(1 + \frac{\alpha}{\omega_{\text{pole1}}^2 - (\omega + i\Gamma)^2} \right. \\ &\quad \left. + \frac{\beta}{\omega_{\text{pole2}}^2 - (\omega + i\Gamma)^2} \right) \\ &= \epsilon_0 \frac{[(\omega + i\Gamma)^2 - \omega_{\text{zero1}}^2][(\omega + i\Gamma)^2 - \omega_{\text{zero2}}^2]}{[(\omega + i\Gamma)^2 - \omega_{\text{pole1}}^2][(\omega + i\Gamma)^2 - \omega_{\text{pole2}}^2]}. \end{aligned} \quad (8)$$

We assume that $\omega_{\text{pole}2} > \omega_{\text{pole}1}$, $\omega_{\text{zero}1}$ and $\omega_{\text{zero}2}$ depend on α and β . Generally, α and β are independent of each other; however, to simplify the discussion we consider the case for which

$$\begin{aligned}\alpha &= \alpha_0(\omega_{\text{pole}2}^2 - \omega_{\text{pole}1}^2) > 0, \\ \beta &= -(\sqrt{\alpha_0} \mp 1)^2(\omega_{\text{pole}2}^2 - \omega_{\text{pole}1}^2) < 0,\end{aligned}\quad (9)$$

so that the two zeros are equal:

$$\omega_{\text{zero}1} = \omega_{\text{zero}2} = \omega_{\text{pole}1} \pm \sqrt{\alpha_0}(\omega_{\text{pole}2}^2 - \omega_{\text{pole}1}^2), \quad (10)$$

where α_0 is a positive real number. The poles and zeros for the upper sign in Eqs. (9) and (10) are shown in Fig. 2. Provided that α_0 is large enough, one may have $(3\pi/4) < |\varphi_1| < \pi$ and $(3\pi/4) < |\varphi_2| < \pi$, such that $(3\pi/2) < \phi_\epsilon < 2\pi$, where φ_1 is the principal value of the argument of $\frac{[(\omega+i\Gamma)^2 - \omega_{\text{zero}1}^2]}{[(\omega+i\Gamma)^2 - \omega_{\text{pole}1}^2]}$ and φ_2 is the principal value of the argument of $\frac{[(\omega+i\Gamma)^2 - \omega_{\text{zero}2}^2]}{[(\omega+i\Gamma)^2 - \omega_{\text{pole}2}^2]}$. Hence, the real part of $\tilde{\epsilon}$ is positive.

Given that $\mu = \mu_0$ is real and positive, the system is a RHM. However, the refractive index will have a negative real part, since $(3\pi/4) < \phi_n = (\phi_\epsilon/2) < \pi$. Similarly, for the lower sign in Eqs. (9) and (10) (not shown in the figure), we can have $-2\pi < \phi_\epsilon < -(3\pi/2)$. Hence, the real part of $\tilde{\epsilon}$ is again positive and the system is also a RHM. The refractive index will also have a negative real part, since $-\pi < \phi_n = (\phi_\epsilon/2) < -(3\pi/4)$. It is interesting to note that for this system the imaginary part of the refractive index is now negative. This indicates that while the system with the upper sign in Eqs. (9) and (10) is lossy, the system with the lower sign has gain.

Numerical estimates show that for certain frequency ranges RHM with negative refractive indices may be realized experimentally. We considered gas-phase as well as condensed-phase media, and here we provide two specific examples using values typical of condensed molecular media. In these examples the number densities are somewhat high; however, the results show that it is plausible that such systems may be realized.

α and β are given by

$$\alpha = \frac{2N_1\bar{p}_1^2\omega_{\text{pole}1}}{\epsilon_0\hbar} \quad \text{and} \quad \beta = \frac{2N_2\bar{p}_2^2\omega_{\text{pole}2}}{\epsilon_0\hbar}, \quad (11)$$

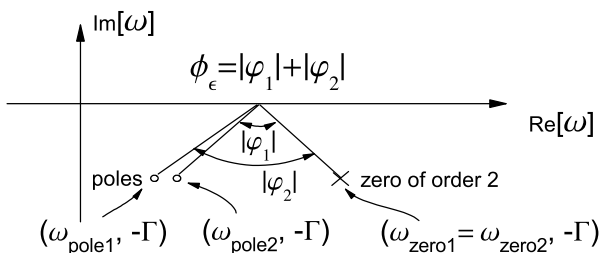


FIG. 2. The zero-pole structure of a RHM that exhibits a negative refractive index.

where \bar{p}_1 and \bar{p}_2 are the transition dipole moments, and N_1 and N_2 the number densities of molecular species 1 and 2, respectively. We now assume that species 1 is in its ground state and has $N_1 = 3 \times 10^{28} \text{ m}^{-3}$ with a resonance at $\omega_{\text{pole}1} = 14000 \text{ cm}^{-1}$. Further, we take $\bar{p}_1 = \bar{p}_2 = 3 \text{ D}$, and $\Gamma = 400 \text{ cm}^{-1}$. If species 2 is in the excited state at $\omega_{\text{pole}2} = 20000 \text{ cm}^{-1}$ with $N_2 = 2.52 \times 10^{27} \text{ m}^{-3}$, then the real part of the refractive index becomes negative between 445 and 530 nm. For the wavelengths between 455 and 495 nm the material is moreover right handed (with $0 < \text{Re}[\tilde{\epsilon}] < 0.6\epsilon_0$ and $\tilde{\mu} = \mu_0$), i.e., the real part of the refractive index dominates, and reaches -1.2 in this wavelength range. The parameters used above are based on the upper sign in Eqs. (9) and (10), and the system is lossy for the frequency range where the real part of the refractive index becomes negative, as can be seen in Fig. 3(a).

Similarly, we can envision a system with gain by considering the lower sign case in Eqs. (9) and (10). Assuming $N_1 = 2 \times 10^{27} \text{ m}^{-3}$ for the noninverted species, $N_2 = 1.75 \times 10^{28} \text{ m}^{-3}$ for the inverted species, $\omega_{\text{pole}1} = 14000 \text{ cm}^{-1}$, $\omega_{\text{pole}2} = 20000 \text{ cm}^{-1}$, $\Gamma = 400 \text{ cm}^{-1}$, and taking $\bar{p}_1 = \bar{p}_2 = 3 \text{ D}$, we find that the real part of the refractive index becomes negative (for the wavelength range 650 to 920 nm), and within the range of 720 to 915 nm, the material is right handed (with $0 < \text{Re}[\tilde{\epsilon}] < 0.65\epsilon_0$ and $\tilde{\mu} = \mu_0$) and its refractive index reaches -1.1 [see Fig. 3(b)]. However, now the imaginary part of the index is also negative (i.e., the system has gain). The need to pump the material may be considered a disadvantage for practical implementations of negative-index media, but may facilitate observation of the effect by compensating absorption.

Our preliminary calculations suggest that right-handed two-component systems with a negative refractive index support evanescent wave amplification predicted in LHM [5], although the underlying details seem to differ.

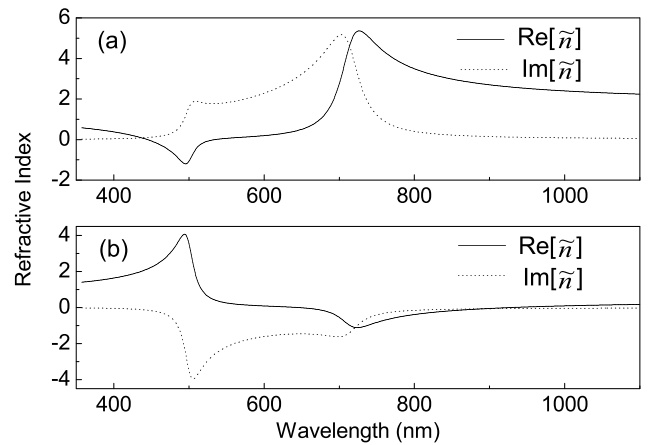


FIG. 3. The real and imaginary parts of the refractive index for two-component systems with loss (a) and gain (b). See text for details.

By considering two-component systems in which one component (molecular species) may be inverted, we predict the existence of nonmagnetic amorphous solids and liquids that have a negative refractive index. Remarkably, the materials are “right handed,” that is to say, the real part of the permittivity and the permeability are both positive at the frequency of interest. Moreover, the two-component media have gain which can overcome the absorption that plagues most “left-handed” negative-index metamaterials (c.f. [27]). Because we make use of “ordinary” resonances in the permittivity, our proposed route to negative refraction opens the possibility of tailoring negative-index media simply by choosing their molecular composition. We expect that this will make it possible to observe negative refractive indices at visible wavelengths in appropriately chosen mixtures.

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