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Investigation of negative refractive index in reciprocal chiral materials

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ABSTRACT

It is well known that there exist both natural materials (such as milk or sugar solution) possessing chiral (or handed) properties, as well as an increasing list of man-made materials (such as sodium bromate) that exhibit chirality. One of the principal properties of chirality is that light of any arbitrary polarization, when propagating through a chiral material, splits up into two circular polarizations propagating in different directions. In the past decade or longer, researchers have investigated electromagnetic transverse (plane) wave propagation across a non-chiral/chiral interface, and determined the electromagnetic Fresnel coefficients for such propagation. Traditionally, such coefficients are derived under the assumption that the transmitted circular polarizations in the chiral material have wave numbers that are numerically positive, and nominally point in the direction of electromagnetic energy flow. However, it turns out that the actual solution for the wavenumbers obtained from applying Maxwell's equations to an unbounded, isotropic chiral material yields four possible values dependent upon the chirality parameter. In this paper, we examine the emergence of these wavenumbers, and thereafter explore the conditions necessary for the resulting field solutions to have counter-propagating energy flow and wave vector. Such conditions, if feasible, represent an environment leading to an effectively negative refractive index being generated within the chiral material. Accordingly, propagation within a chiral medium through the mechanism of negative refractive indices may be studied in order to better understand the corresponding optical properties of such materials vis-a-vis transmission of an electromagnetic wave into and out of such a region. The results obtained may be applied to compare negative index chiral materials with the broader emerging field of negative index metamaterials, and explore possible applications.

Keywords: Chirality; handed media; polarization; constitutive relations; chiral admittance; wavenumber; Poynting vector; negative index

1. INTRODUCTION

Chiral or gyrotropic media possess handedness (i.e. dominant right- or left-circular polarization). The word chiral is derived from the Greek word, *chiro*, implying the hand. A chiral object has a non-superimposable mirror image whereas an achiral object has an identical (superimposable) mirror image. Chirality or handedness is observed frequently in nature; sea shells, spirals, bacteria and a host of organic and inorganic materials exhibit varied proportions of right- or left-handedness. An electromagnetic wave propagating through a chiral medium undergoes a change in polarization depending upon its initial polarization, direction of propagation, and other properties. Examples of chiral media include quartz, metal helices in epoxy, copper strings in dielectric and ferroelectric ceramics.

More recently, researchers from a variety of disciplines such as biology¹ and chemistry^{2,3} have published results which have a strong relevance to electromagnetics and optics applications of chirality. It is well known now that many crystals such as cinnabar, sodium chlorate, sodium bromate and liquids such as sugar solution, corn syrup and turpentine exhibit optical activity. Various groups in the United States^{4,5}, Europe and Russia⁶⁻⁸ have been working on chiral media and their applications.

The terminology and notations used in discussions about chiral media vary. The term *chirality* was coined by Kelvin⁹, defining this concept as a property of an object that is not superimposable to its mirror image. In the past, electromagnetic chirality was studied primarily by researchers in optics. Hence, chiral media are commonly called

optically active media. Incidentally, researchers in Russia describe such materials gyrotropic media. More recently, both Russian and English language journals have adopted the name *chiral* for such a medium.

In general, the analysis of chiral media (which are generally isotropic, but may possess reciprocal or nonreciprocal properties- the latter a subject of considerable controversy^{8,10,11}) involves highly complex mathematics, including the application of Green's functions, and a variety of special transforms. Since the basic constitutive relations in the Maxwell's equations are modified in the chiral media to include terms containing so-called chiral and reciprocity parameters (κ and χ respectively), a relatively simple method was developed by Banerjee *et.al*¹², whereby solutions for the vector electric field in an unbounded chiral medium are obtained via a dual transform technique. The technique assumes paraxiality and slowly varying envelopes, and proceeds by applying successively a 2-D (transverse) spatial Fourier transform and a Laplace transform along the propagational or the longitudinal direction. A straightforward application of the above technique has been shown to confirm that any arbitrary polarized uniform plane wave transforms into two circular polarizations upon propagation through the chiral medium.

The focus of this paper is on the question of wave propagation in an isotropic, reciprocal, and unbounded chiral medium. We apply the Maxwell curl equations to such a medium, and use the corresponding constitutive relations to obtain a set of field solutions for a given propagation vector. Using the Poynting vector corresponding to these solutions, conditions are then examined that lead to a net Poynting vector or energy flow *opposite* in direction to the propagation vector. When these conditions are satisfied, it is surmised that the resulting chiral medium constitutes an environment of negative refractive index- a subject of considerable current interest. The investigation of negative index materials extends to a variety of media, and the phenomenon is often viewed in terms of dichroic behavior in orthorhombic and other materials whose electric permittivity and magnetic permeability exhibit negative real parts¹³⁻¹⁵. We show further that the net propagating field in the chiral medium consists of an elliptical polarization resulting from the superposition of two (left- and right-) circular polarizations of (generally) dissimilar amplitudes. Finally, we speculate that in general a negative index chiral material requires a relatively large magnitude of the chirality parameter (not typical of conventional chiral materials with positive indices).

2. PLANE WAVE PROPAGATION IN AN UNBOUNDED, ISOTROPIC AND RECIPROCAL CHIRAL MEDIUM

2.1 Definition of the problem

We consider arbitrary electromagnetic wave propagation in an unbounded, isotropic and reciprocal ($\chi = 0$) chiral medium. Maxwell's equations, along with the modified constitutive relations appropriate for the chiral medium are then used to determine the resulting relationships between the field components.

2.2 Solutions using phasor Maxwell equations and constitutive relations

The chiral non-reciprocal medium is described by the constitutive relations (usually in the frequency domain)

$$\bar{D} = \epsilon \bar{E} + \xi \bar{H} \quad , \quad (1)$$

$$\bar{B} = \zeta \bar{E} + \mu \bar{H} \quad , \quad (2)$$

where,

$$\xi = -j \kappa \sqrt{\mu_0 \epsilon_0} \quad ,$$

$$\zeta = \xi^* = j \kappa \sqrt{\mu_0 \epsilon_0} \quad ,$$

μ_0, ε_0 are the permeability and permittivity of free space, and κ is the so-called chirality parameter, which is dimensionless. We may note that the chirality parameter described above is related to the so-called chirality admittance γ that appears elsewhere⁵ through:

$$\gamma = -\frac{\kappa \sqrt{\mu_0 \varepsilon_0}}{\mu} .$$

and ε (to appear later) and μ are the nominal electric permittivity and magnetic permeability parameters associated with the chiral material. The chirality parameter κ defines the degree of handedness of the material. In a racemic mixture where the chiral materials of both types, i.e., right- and left-handed, are equal, and for non-chiral materials the chirality parameter κ goes to zero. The j emphasizes the frequency domain nature of these relations, and the harmonic convention $e^{j\omega t}$ is implied.

Next, we consider the Maxwell curl equations in the phasor domain written in the form:

$$\begin{aligned} \bar{k}_0 \times \bar{E} &= \omega [\zeta \bar{E} + \mu \bar{H}] , \\ \bar{k}_0 \times \bar{H} &= \omega [\varepsilon \bar{E} + \xi \bar{H}] , \end{aligned} \quad (3a,b)$$

where \bar{k}_0 is the wave vector, ω is the (monochromatic) wave frequency, and \bar{E} and \bar{H} are the electric and magnetic field intensities, respectively.

The solutions for the above fields are obtained more readily by assuming a wave vector pointed in the Z-direction (which is arbitrary, and hence general, in an unbounded medium). This approach simplifies the resulting characteristic matrix to a 4×4 instead of a 6×6 . Using such a wave vector, it is simple to show that the longitudinal components of the fields vanish, resulting in a purely *transverse* propagation in the bulk chiral material. The following set of homogeneous equations for the field components is then obtained from eqs. (3a,b):

$$\begin{aligned} j \alpha E_x + k_{0z} E_y + \omega \mu H_x &= 0 , \\ k_{0z} E_x - j \alpha E_y - \omega \mu H_y &= 0 , \\ \omega \varepsilon E_x - j \alpha H_x - k_{0z} H_y &= 0 , \\ \omega \varepsilon E_y + k_{0z} H_x - j \alpha H_y &= 0 , \end{aligned} \quad (4a-d)$$

where the parameter $\alpha = \omega \kappa \sqrt{\mu_0 \varepsilon_0}$, and has the dimension m^{-1} , and k_{0z} is the wavenumber.

Finding non-trivial solutions for the homogeneous eqs.(4a-d) requires that the determinant of the coefficient matrix must vanish. This leads to the well-known condition for the wavenumber in the chiral medium:

$$k_{0z} = \pm \omega (\kappa \sqrt{\mu_0 \varepsilon_0} \pm \sqrt{\mu \varepsilon}) , \quad (5)$$

which indicates a set of four possible values of the wavenumber that satisfy the non-trivial field solutions. We may note that the k_{0z} values depend on the chirality parameter κ .

We next assume that the field component E_x is known. From the homogeneous equations (4a-c), assuming the known E_x value, the field solutions are obtained after some algebra as follows:

$$\begin{aligned}
& E_x , \\
& E_y = \frac{-\omega^2 \mu \varepsilon + \alpha^2 + k_{0z}^2}{2 j \alpha k_{0z}} E_x , \\
& H_x = \frac{-k_{0z}^2 + \omega^2 \mu \varepsilon + \alpha^2}{2 j \alpha \omega \mu} E_x , \\
& H_y = \frac{-\alpha^2 + k_{0z}^2 + \omega^2 \mu \varepsilon}{2 \omega_0 \mu k_{0z}} E_x .
\end{aligned} \tag{6a-d}$$

An important observation may be made here in view of the above solutions. Note that these solutions are true for an unbounded medium (and may be readily shown to satisfy the Maxwell curl equations), and are general in the sense that the Z-directed wave vector is essentially arbitrary in an unbounded medium. We may also observe that the transverse electric field components have a phase difference of $\pm \pi / 2$, but are unequal in amplitude. Such a condition arises from the superposition of a left- and a right-circular wave that have unequal amplitudes. Hence, the total electric field in the chiral medium (regardless of positive or negative index) is *elliptically polarized* with a phase difference of $\pm \pi / 2$, and is made up of two circular polarization (LCP and RCP), as is well known.

3. POYNTING VECTOR AND WAVE VECTOR

Next, based on the above field solutions, we obtain the corresponding Poynting vector to determine the nature of power flow through the chiral material. Since the field turns out to be transverse to the Z-direction, it is clear that the Poynting vector will be longitudinal. The intention here is to find the conditions under which the Poynting and the wave vector may be counter-propagating.

3.1 Complex Poynting vector

Using the field solutions (eqs. 6a-d), the Poynting vector is obtained after considerable algebra as follows:

$$\overline{E} \times \overline{H}^* = \frac{-3\alpha^4 + 2\alpha^2 k_{0z}^2 + 2\alpha^2 \omega^2 \mu \varepsilon + k_{0z}^4 + \omega^4 \mu^2 \varepsilon^2 - 2\omega^2 \mu \varepsilon k_{0z}^2}{4\alpha^2 \omega \mu k_{0z}} |E_x|^2 \hat{a}_z . \tag{7}$$

We next make the following observations. Since the Poynting vector is in the longitudinal direction (as is the wave vector), a positive value of k_{0z} requires the numerator in eq.(7) to be positive in order for the power to be transported in the same direction as the propagation vector (or, equivalently, the phase velocity vector). Such a scenario would then correspond to the usual energy transport in the chiral medium under an equivalent positive refractive index. In most cases, such a scenario is shown to be satisfied by the choice of the “+” sign inside the parentheses of eq.(5), leading to the two well-known conventional wave numbers. However, one may mathematically construct various scenarios under which the power flow in eq.(7) and the wave vector may be counter-propagating. One may, for example, examine this in terms of possible negative real values of the parameters μ and/or ε (as is done for the dichroic problem¹⁴), or, alternatively, consider possible effects of the chirality parameter κ itself. We pursue one such condition in the next section.

3.2 Condition for the counterpropagation of power flow and wave vector

From eq.(7), we consider the case where the numerator is negative for a given negative value of the wavenumber k_{0z} , such that the wave vector of the field and the power transport may be in opposite directions. Requiring the numerator of eq.(7) to be negative leads to:

$$3\omega^4 \kappa^4 \mu_0^2 \varepsilon_0^2 + 2\omega^2 \mu \varepsilon k_{0z}^2 > 2\omega^2 \kappa^2 \mu_0 \varepsilon_0 k_{0z}^2 + 2\omega^4 \kappa^2 \mu_0 \varepsilon_0 \mu \varepsilon + k_{0z}^4 + \omega^4 \mu^2 \varepsilon^2, \quad (8)$$

which, after substituting eq.(5), and some algebra leads to the condition:

$$\pm \kappa \sqrt{\mu_0 \varepsilon_0} > -\sqrt{\mu \varepsilon}. \quad (9)$$

The above condition leads to the two cases, (a) $\kappa < -\sqrt{\mu_r \varepsilon_r}$, and (b) $\kappa > \sqrt{\mu_r \varepsilon_r}$, where μ_r and ε_r are the relative permeability and permittivity of the chiral material, respectively. In general, the above result may be written as:

$$|\kappa| > \sqrt{\mu_r \varepsilon_r}. \quad (10)$$

We remark that these conditions are identical to those observed recently in the literature^{16,17}, derived via the route of carrying out the dot product between the propagation vector and the complex Poynting vector.

4. CONCLUSIONS

From eq.(10), we may conclude that in general, one requires a chiral material with a relatively large chirality parameter in order to reasonably expect an effective negative refractive index. Typical chiral materials commonly exhibit smaller values of the chirality parameter (described as *weak chirality* in the literature). We also note that the condition specified by eq.(9) offers a positive and negative regime of κ (both of large magnitude) in which a negative index condition may be realized¹⁸. Another parallel effort involves the examination of nonlinear propagation in negative index materials in general¹⁹. A possible material in the physical world where such behavior may be manifested is the DNA helix, where the orientation of the spiral may lead to one or the other condition being satisfied. The analysis carried out here shows the general elliptical nature of wave polarization in a chiral material, characterized by two (LCP and RCP) circular polarizations of unequal amplitudes. In propagating through a non-chiral and chiral interface, these two polarizations generally assume two different spatial directions and phase velocities for the case of positive index materials. The corresponding case for negative index materials, and the associated Fresnel coefficients (both amplitude and energy) will be explored further.

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