Progress In Electromagnetics Research Letters, Vol. 52, 57–62, 2015

Rigorous Approach of the Constitutive Relations for Nonlinear Chiral Media

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Abstract—A new mathematical approach is proposed to highlight the nonlinear effect in a chiral medium, which is due to the magnetization vector under the influence of a strong electric field. In *a chiral media, one can notice the coupling between the electric and magnetic quantities, which appears in the constitutive relations of the medium.* According to our proposed approach, we illustrate the existence of the difference between a nonlinear achiral medium and a nonlinear chiral medium where not only the polarization vector has a nonlinear form but also the magnetization vector. Thus, the nonlinear chiral medium is described by the new constitutive relations $\vec{D} = \varepsilon_g \vec{E} + \sqrt{\varepsilon_0 \mu_0} \xi_{EH} \vec{H}$ and $\vec{B} = \mu \vec{H} + \sqrt{\varepsilon_0 \mu_0} \xi_{HE}^{g} \vec{E}$. Therefore, a better fundamental understanding of the interaction between the electromagnetic waves and chiral media can be contemplated.

1. INTRODUCTION

Materials with intrinsic properties can combine several functionalities at the same time, which allows for chiral materials to be introduced recently in various scientific disciplines and to be used adequately in nanotechnology.

In a chiral material, the physical behavior is a consequence of chirality. The establishment of the relationships between physical and geometrical concepts emerged during the 19th century and the beginning of the 20th century, where the concepts of chirality and optical activity were definitively related. Main progress in the field of the chirality and the optical activity during the last century and the beginning of this century is attributed to A. Fresnel, J. B. Biot and L. Pasteur. In the beginning, it was in 1809 that Malus observed the existence of two images when light was refracted through a birefringent crystal [1]. More recently, in 1920 and 1922, Lindman devised a macroscopic (molar) model of the phenomenon by using microwaves instead of light, and wire spirals instead of chiral molecules [1]. The physicists and the chemists then developed theories with the aim of explaining and quantifying the phenomenon of optical activity.

The optical activity is the property, which has a chiral structure, to interact with an electromagnetic radiation. It manifests itself through the existence of optical activity, optical phenomenon of dispersion, circular dichroism and circular polarization of emission [2–4].

The invention of lasers in 1960 invalidated some part of the approximations hitherto used in optics. In fact, the lasers involved such powers that the magnitude of the electrons cohesion energy in the atoms (or the molecules) is reached. The linear approximation is not then valid anymore, and it is necessary to call upon nonlinear optics. We can report some of the phenomena with nonlinear traditional models (Kerr effect, mixture with three or four waves, etc.) [5].

The study of a chiral medium exhibits very great complexity and requires more rigorous modeling. In this context, several theoretical studies have been conducted on the nonlinearity of the chiral medium

Received 11 February 2015, Accepted 4 March 2015, Scheduled 16 March 2015
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with consideration of the effect of strong electric field generated by the laser on the vector of polarization, without paying attention to the existence of a difference between a chiral medium and an achiral environment, where one must take into account the influence of strong electric field on the vector of magnetization because the macroscopic model of the interaction of electromagnetic wave with chiral structures shows that the magnetization vector is of the following form: $\vec{M} = \varepsilon_0 \chi_m \vec{H} + \chi_{me} \vec{E}$ which is a function of the magnetic and electric fields [1]. With these observations, we propose a new mathematical approach to the constitutive relations of a nonlinear chiral medium, more accurate and in accordance with the physical characteristics of chiral medium, that is what will be presented in this article.

2. FORMULATION

For a few years, research works have actively considered new types of heterogeneous absorbent materials: chiral materials. The chiral material consists of a random dispersion of inclusions in a polymeric or ceramic matrix. Lindman in 1920 and Pickering in 1945 studied in particular the interaction of an electromagnetic wave with a collection of metal helices of the same enantiomorphs form distributed randomly. They then observed a rotation of the electromagnetic wave polarization plane after interaction with the helices.

In 1979, Jaggard et al. [1] presented a macroscopic model of the interaction of an electromagnetic wave with chiral structures. They showed that in the case of a harmonic excitation ($e^{-j\omega t}$) of a chiral medium composed of a collection of metal helices with the same enantiomorphs form drowned in a dielectric matrix, the polarization vector $\vec{P}$ and magnetizing vector $\vec{M}$ can be written as follows

$$\vec{P} = \varepsilon_0 \chi_e \vec{E} + \chi_{em} \vec{H} \tag{1}$$

$$\vec{M} = \varepsilon_0 \chi_m \vec{H} + \chi_{me} \vec{E} \tag{2}$$

χ_e: Electric susceptibility, χ_m: Magnetic susceptibility, χ_{em} and χ_{me}: The cross-susceptibilities. ε_0: Permittivity of free space.

$\vec{E}$ and $\vec{H}$ are the electric- and magnetic-fields, respectively.

The helical model focused thinking on the physical aspects of optical activity seeing the role of electric and magnetic dipoles, their phase relations, and the importance of reflection symmetries. Amusingly, the simple figure of such a helical model was a great success at presentations [4] (see Fig. 1).

Thus the vectors of polarization and magnetizing have the following forms:

$$\vec{P} = \vec{P}_e + \vec{P}_m \tag{3}$$

$$\vec{M} = \vec{M}_m + \vec{M}_e \tag{4}$$

$$\vec{P}_e = \varepsilon_0 \chi_e \vec{E} \tag{5}$$

$$\vec{P}_m = \chi_{em} \vec{H} \tag{6}$$

$$\vec{M}_e = \chi_{me} \vec{E} \tag{7}$$

$$\vec{M}_m = \varepsilon_0 \chi_m \vec{H} \tag{8}$$

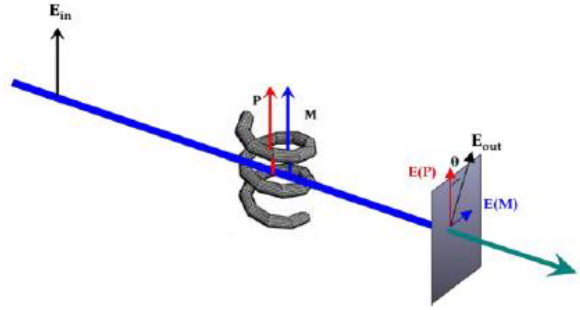

Figure 1. An intuitive view of (the optical activity in) a helical molecule [4].

where $\vec{P}_e$ and $\vec{P}_m$ are the polarizations induced in the chiral medium by $\vec{E}$ and $\vec{H}$ respectively. The same is for: magnetizing $\vec{M}_e$ and $\vec{M}_m$

3. NONLINEAR CHIRAL EFFECT

The description of such effects is based on an extension of the concept of linear propagation of the electromagnetic field in the matter. It rests on the use of the macroscopic Maxwell equations in which polarization (magnetizing) is expressed by means of a development in powers of amplitude of the fields present in the medium. In the same way, the linear properties of the chiral medium are described by means of only one quantity: linear susceptibility, whereas the nonlinear properties of a material are characterized by a certain number of susceptibilities expressing the nonlinearity [3, 4]. In the linear chiral media $\vec{P}$ and $\vec{M}$ are directly proportional to $\vec{E}$ and $\vec{H}$ respectively. Therefore, the principle of superposition applies. The oscillating dipoles at the electromagnetic wave frequency will radiate a field of the same frequency in the medium, and thus will modify the optical wave propagation [3–5]. However, when the field is sufficiently intense, about the inter-atomic field, the response of the chiral medium (thus polarization, magnetizing) is a nonlinear function of the excitation. A laser wave corresponds to an electromagnetic field oscillating at a frequency of about 10^{13} to 10^{15} Hz. Under the action of the electric field of such a wave, the dielectric electric charges are subjected to an oscillating movement of the same frequency, forming a set of oscillating dipoles [3–5]. The effect of the magnetic field on the charged particles is much weaker and can be neglected.

Using known approximations, the self-susceptibilities can be written as

$$\delta_e = \left((2l)^2 C/\varepsilon_0\right) \tag{9}$$

$$\delta_m = \left(\left(\pi a^2\right)^2 \mu_0/L\right) \tag{10}$$

where C and L are, respectively, the capacitance and the inductance of the body, and $2l$ and $2a$ represent the length and the width of the short helix (Fig. 2) [1]. It can be shown that the cross-susceptibilities might be given by [1]:

$$\delta_{em} = \delta_m \left(2l/\pi a^2 k\right) \tag{11}$$

$$\delta_{me} = \delta_e \left(\pi a^2 k/2l\right) \tag{12}$$

$\vec{k}$: Wave vector.

Eq. (12) expresses the relation between cross-susceptibility χ_{me} and electric susceptibility χ_e. This remark illustrates the fact that we cannot neglect the nonlinear magnetizing [4], which is responsible for important phenomena in a nonlinear chiral medium.

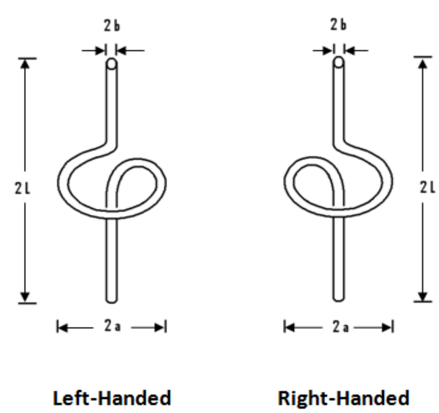

Figure 2. Idealized short helices used in calculations [1].

The model of material under study may give place to a certain number of nonlinear chiral effects. For these reasons the correspondence between the demonstration of these effects and the formulation of corresponding susceptibilities must allow a total and universal description of nonlinear chiral effects. The total polarization and total magnetizing of the nonlinear chiral medium will be then both formulated according to a development in Taylor series as follows:

$$\vec{P} = \chi_{em}^{(1)}\vec{H} + \varepsilon_0\chi_e^{(1)}\vec{E} + \varepsilon_0\chi_e^{(2)}\vec{E}\vec{E} + \varepsilon_0\chi_e^{(3)}\vec{E}\vec{E}\vec{E} + \dots \tag{13}$$

$$\vec{M} = \varepsilon_0\chi_m^{(1)}\vec{H} + \chi_{me}^{(1)}\vec{E} + \chi_{me}^{(2)}\vec{E}\vec{E} + \chi_{me}^{(3)}\vec{E}\vec{E}\vec{E} + \dots \tag{14}$$

where Eq. (15) and Eq. (16) represent linear polarization and linear magnetizing, respectively,

$$\vec{P}_e^L = \varepsilon_0\chi_e^{(1)}\vec{E} \tag{15}$$

$$\vec{M}_e^L = \chi_{me}^{(1)}\vec{E} \tag{16}$$

And Eq. (17), Eq. (18), the expressions representative of nonlinear polarization and nonlinear magnetizing:

$$\vec{P}_e^{NL} = \varepsilon_0\chi_e^{(2)}\vec{E}\vec{E} + \varepsilon_0\chi_e^{(3)}\vec{E}\vec{E}\vec{E} + \dots \tag{17}$$

$$\vec{M}_e^{NL} = \chi_{me}^{(1)}\vec{E} + \chi_{me}^{(2)}\vec{E}\vec{E} + \chi_{me}^{(3)}\vec{E}\vec{E}\vec{E} + \dots \tag{18}$$

with the terms $\chi_e^{(n)}$ and $\chi_{me}^{(n)}$ expressing macroscopic susceptibilities of the nonlinear chiral medium. They are also called the nonlinear susceptibilities of order (n) and represented by tensors of order $(n+1)$.

The nonlinear polarization, given by [3–6], is expressed according to an algebraic development of tensorial products between components. This is what allowed us to reformulate the expressions Eqs. (17) and (18) in the following forms:

$$P_{(e)i}^{NL} = \varepsilon_0\sum_{j,k}\chi_{(e)ijk}^{(2)}E_j(\omega_1)E_k(\omega_2) + \varepsilon_0\sum_{j,k,l}\chi_{(e)ijkl}^{(3)}E_j(\omega_1)E_k(\omega_2)E_l(\omega_3) + \dots \tag{19}$$

$$M_{(e)i}^{NL} = \sum_{j,k}\chi_{(me)ijk}^{(2)}E_j(\omega_1)E_k(\omega_2) + \sum_{j,k,l}\chi_{(me)ijkl}^{(3)}E_j(\omega_1)E_k(\omega_2)E_l(\omega_3) + \dots \tag{20}$$

where: $1 \le i, j, k \le 3$.

4. CONSTITUTIVE RELATIONS OF NONLINEAR CHIRAL MEDIA

The traditional laws of electromagnetism impose that:

$$\vec{D} = \varepsilon_0\vec{E} + \vec{P} \tag{21}$$

$$\vec{B} = \mu_0\left(\vec{H} + \vec{M}\right) \tag{22}$$

where $\vec{D}$ and $\vec{B}$ are the electric displacement and the magnetic flux density, respectively.

Referring to articles [6–10], we can deduce from Eq. (13) and Eq. (14) the constitutive equations of the nonlinear chiral medium according to the following forms:

$$\vec{D} = \varepsilon\vec{E} + \sqrt{\varepsilon_0\mu_0}\xi_{EH}\vec{H} + \vec{P}_e^{NL} \tag{23}$$

$$\vec{B} = \mu\vec{H} + \sqrt{\varepsilon_0\mu_0}\xi_{EH}^*\vec{E} + \mu_0\vec{M}_e^{NL} \tag{24}$$

where ε and μ are respectively the permittivity and permeability of the chiral medium.

The linear coefficients of chirality are given by [6, 7]:

$$\xi_{EH} = \gamma - j\kappa \tag{25}$$

$$\xi_{HE} = \xi_{EH}^* \tag{26}$$

where γ is the non-reciprocity parameter and κ the linear chirality parameter.

Various possible writings describing coupling between the electric and magnetic quantities lead to various shapings of the formalisms for the chiral media description. Generally, by employing the formalism of Shivola and Lindell [6, 7], our constitutive equations of a nonlinear chirality medium will be expressed as follows

$$\vec{D} = \varepsilon_g \vec{E} + \sqrt{\varepsilon_0 \mu_0} \xi_{EH} \vec{H} \tag{27}$$

$$\vec{B} = \mu \vec{H} + \sqrt{\varepsilon_0 \mu_0} \xi^g_{HE} \vec{E} \tag{28}$$

$$\varepsilon_g = \varepsilon + \varepsilon_{NL} \tag{29}$$

$$\xi^g_{HE} = \xi^*_{EH} + \xi^{NL}_{HE} \tag{30}$$

where ε_{NL} is the nonlinear permittivity, while ξ^g_{HE} and ξ^{NL}_{HE} are the total chirality coefficient and nonlinearity coefficient.

5. PROPAGATION IN A NONLINEAR CHIRAL MEDIUM

Maxwell's equations are given by:

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \tag{31}$$

$$\vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t} \tag{32}$$

Substituting Eq. (28) in Eq. (31) and Eq. (27) in Eq. (32) yields

$$\vec{\nabla} \times \vec{E} = -\mu \frac{\partial \vec{H}}{\partial t} - \sqrt{\varepsilon_0 \mu_0} \frac{\partial \xi^g_{HE} \vec{E}}{\partial t} \tag{33}$$

$$\vec{\nabla} \times \vec{H} = \frac{\partial \varepsilon_g \vec{E}}{\partial t} + \sqrt{\varepsilon_0 \mu_0} \xi_{EH} \frac{\partial \vec{H}}{\partial t} \tag{34}$$

Then taking the *curl* of both sides of Eq. (33) results in

$$\vec{\nabla} \times \vec{\nabla} \times \vec{E} = -\mu \frac{\partial \vec{\nabla} \times \vec{H}}{\partial t} - \sqrt{\varepsilon_0 \mu_0} \frac{\partial \vec{\nabla} \times \left(\xi^g_{HE} \vec{E} \right)}{\partial t} \tag{35}$$

$$\nabla^2 \vec{E} = \mu \frac{\partial \vec{\nabla} \times \vec{H}}{\partial t} + \sqrt{\varepsilon_0 \mu_0} \frac{\partial \vec{\nabla} \times \left(\xi^g_{HE} \vec{E} \right)}{\partial t} \tag{36}$$

By substituting Eq. (34) in Eq. (36) we obtain:

$$\nabla^2 \vec{E} = \mu \frac{\partial^2 \left(\varepsilon_g \vec{E} \right)}{\partial t^2} + \mu \sqrt{\varepsilon_0 \mu_0} \xi_{EH} \frac{\partial^2 \vec{H}}{\partial t^2} + \sqrt{\varepsilon_0 \mu_0} \frac{\partial \vec{\nabla} \times \left(\xi^g_{HE} \vec{E} \right)}{\partial t} \tag{37}$$

Then expressing $\vec{H}$ with respect to $\vec{E}$ from Eq. (33) and substituting in Eq. (37) gives:

$$\nabla^2 \vec{E} = \mu \frac{\partial^2 \left(\varepsilon_g \vec{E} \right)}{\partial t^2} - \sqrt{\varepsilon_0 \mu_0} \xi_{EH} \frac{\partial \vec{\nabla} \times \vec{E}}{\partial t} - \varepsilon_0 \mu_0 \xi_{EH} \frac{\partial^2 \left(\xi^g_{HE} \vec{E} \right)}{\partial t^2} + \sqrt{\varepsilon_0 \mu_0} \frac{\partial \vec{\nabla} \times \left(\xi^g_{HE} \vec{E} \right)}{\partial t} \tag{38}$$

In this section, we inferred the cubic nonlinear Schrödinger equation for a chiral medium term with a nonlinear term of magnetizing. This result is a generalization of the work done by the authors in [11–15].

6. CONCLUSION

Our research work is concerned with a new formulation of the constitutive relation relating to the magnetic effect, in order to more rigorously model the physical nature of the chiral effects and to generalize the main macroscopic models of Chirality to nonlinear effects. In prospects, it will be thus very important to numerically solve the nonlinear equation, where the soliton propagation rises from this effect which balances the effects of the nonlinearity of the interaction and material dispersion.

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