Spatial dispersion and energy in strong chiral medium

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Abstract: Since the discovery of backward-wave materials, people have tried to realize strong chiral medium, which is traditionally thought impossible mainly for the reason of energy and spatial dispersion. We compare the two most popular descriptions of chiral medium. After analyzing several possible reasons for the traditional restriction, we show that strong chirality parameter leads to positive energy without any frequency-band limitation in the weak spatial dispersion. Moreover, strong chirality does not result in a strong spatial dispersion, which occurs only around the traditional limit point. For strong spatial dispersion where higher-order terms of spatial dispersion need to be considered, the energy conversation is also valid. Finally, we show that strong chirality need to be realized from the conjugated type of spatial dispersion.

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References and links

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1. Introduction

Chirality is first referred to a kind of asymmetry in geometry and group theory in mathematics. The asymmetry exists broadly in organic molecules, crystal lattices, and liquid crystals, leading to two stereoisomers, dextrorotatory and laevorotatory, as a hot research domain in stereochemistry. If the two stereoisomers coexist in one molecule (mesomer), or equally different stereoisomers get mixed (raceme), there will be no special characters other than the common magneto-dielectric. When we get one pure stereoisomer, however, interesting phenomena occur with an incident linearly-polarized wave, which can be seen as a superposition of two dual circularly-polarized waves. In case of perpendicular incidence, the two different circularly polarized waves have different phase velocities and their polarized planes rotate oppositely. As a result, the output polarization direction gets rotated, also known as optical activity or natural optical rotation phenomenon, which was first observed by Arago in 1811. For oblique incidence, the two different polarized waves will split even the medium is isotropic, which was verified by Fresnel using prism series made from dextrorotatory and laevorotatory quartz [1-4]. Moreover, in elementary particle physics, chirality and asymmetry also play important roles, but they are out of the range of this paper.

2. Two major electromagnetic models to describe a chiral medium

The electromagnetic theoretical explanation of optical activity is spatial dispersion [5-8]. Usually, under the weak spatial dispersion, we use the first order (linear) approximation, which is written as

\[
\vec{D} = \varepsilon_{DBF} \vec{E} + \varepsilon_{DBF} \beta \nabla \times \vec{E},
\]

\[
\vec{B} = \mu_{DBF} \vec{H} + \mu_{DBF} \beta \nabla \times \vec{H}.
\]

Such a representation is named as Drude-Born-Fedorov (DBF) relation for a natural result of linearly spatial dispersion. Rotation terms are added to the basic constitutive relation, standing for the spatial dispersion, whose coefficients \( \varepsilon_{DBF} \beta \) or \( \mu_{DBF} \beta \) can be either positive or negative for two stereoisomer structures. Solving the constitutive relation together with Maxwell’s equations, we can easily get two eigenwaves, which are left and right circularly polarized with different wavevectors. There are also some other representations, among which the most common one is deduced by Pasteur and Tellegen as

\[
\vec{D} = \varepsilon \vec{E} + (\chi + i\kappa) \vec{H},
\]

\[
\vec{B} = \mu \vec{H} + (\chi - i\kappa) \vec{E},
\]

in which electromagnetic coupling terms are added to the basic terms. Bi-isotropy or bi-anisotropy is used for calling such constitutive equations, according to the parameters to be scalars or tensors. If \( \kappa = 0 \) and \( \chi \neq 0 \), it is the Tellegen medium; if \( \chi = 0 \) and \( \kappa \neq 0 \), as the
requirement of reciprocity, it is the Pasteur medium:
\[ \vec{D} = \varepsilon \vec{E} + i\kappa \vec{H}, \]  
\[ \vec{B} = \mu \vec{H} - i\kappa \vec{E}. \]  

We pay more attention to such a chiral medium. Positive and negative \( \kappa \) values differentiate two conjugated stereoisomer structures. We assume \( \kappa > 0 \) in the following analysis.

Actually, the constitutive relations above are essentially equivalent, with corresponding parameters to be \[ \varepsilon_{DBF} = \varepsilon \left( 1 - \frac{\kappa^2}{\mu \varepsilon} \right), \]  
\[ \mu_{DBF} = \mu \left( 1 - \frac{\kappa^2}{\mu \varepsilon} \right), \]  
\[ \beta = \frac{\kappa}{\omega (\mu \varepsilon - \kappa^2)}. \]  

It is clear that the parameters are different in such two representations. Then a question may rise up: which are the “true” material permittivity and permeability? The answer is, both. The concepts of permittivity and permeability are effective coefficients derived from a mathematical model. We actually have different mathematical models describing the same physical material. Thus there are different effective parameters describing the proportion of \( \vec{D} \) to \( \vec{E} \) and \( \vec{B} \) to \( \vec{H} \). The rotation terms in DBF model include both real and imaginary parts, resulting in a change in the real part and creating the imaginary chiral terms in the Pasteur model, vice versa. In other words, the difference in representations of coupling terms lead to different permittivity and permeability formulations.

It should be noticed that Faraday gyratory medium can also lead to optical rotation within the plasma or ferrite under an additional DC magnetic field \[9, 10\]. Hence it is not natural, and is usually referred as “gyratory”, “Faraday optical rotation”, “magneto-optical effect”, etc. However, sometimes people do not differentiate “chiral” and “gyratory”. We need pay attention that such two types of optical rotation have different essence and different characters \[10\]. Only natural optical activity is discussed here.

3. Energy and spatial dispersion in strong chiral medium

There is a long dispute on strong chiral medium since it was introduced theoretically \[11\]. Traditional electromagnetic conclusions have limited us to understand strong chirality, i.e. \( \kappa^2 > \mu \varepsilon \[7, 11\] \), until we see the fact that artificial Veselago’s medium \[12\] was successfully realized in certain frequency bands \[13\]. Hence, we have to ask the following question: can strong chiral medium exist?

In Ref. \[8\[11\], the reason for traditional restriction of chirality parameters was concluded as: 1) The wavevector of one eigenwave will be negative; 2) The requirement of a positive definite matrix to keep positive energy:

\[ \begin{bmatrix} \varepsilon & i\kappa \\ -i\kappa & \mu \end{bmatrix}. \]  

With the exploration of backward-wave medium, we know that negative wavevector, or opposite phase and group velocities, are actually realizable. And there is an unfortunate mathematical error in the second reason: in linear algebra, only if it is real and symmetric, positive definite matrix is equivalent to that all eigenvalues should be positive. The matrix \[10\] is a complex one, making the analysis on restriction of positive energy meaningless.
Actually, in a strong bi-isotropic medium with constitutive relations as Eqs. (5) and (6), the energy can be drawn as

\[
w = w_e + w_m
\]

\[
= D \cdot E / 2 + B \cdot H / 2
\]

\[
= \varepsilon |E|^2 / 2 + i\kappa H \cdot E + \mu |H|^2 / 2 - i\kappa E \cdot H
\]

\[
= \varepsilon |E|^2 / 2 + \mu |H|^2 / 2. \tag{11}
\]

Even if the strong bi-isotropic medium is not frequency dispersive, i.e. \( \kappa^2 > \mu \varepsilon \) for whole frequency range, the energy will still keep positive as long as the permittivity and permeability are positive, under the weak spatial dispersion condition. This is quite different from the Veselago’s medium since there is no bandwidth limitation and the frequency dispersive resonances are no longer required. In another word, the strong chiral medium does not contradict the energy conservation, at least in the weak spatial dispersion model.

Therefore, the real reason for traditional strong-chirality limitation is neither negative wavevector nor energy conversation. Next we will point out two other important reasons.

First, with the assumption that \( \varepsilon > 0, \mu > 0, \kappa > 0 \) and \( \kappa > \sqrt{\mu \varepsilon} \), we easily show that \( \varepsilon_{DBF}, \mu_{DBF} \) and \( \beta \) turn to negative from the transformation between Pasteur constitutive relations and DBF relations shown in Eqs. (7)-(9). This is absolutely unacceptable before people realizing Veselago’s medium. Actually, strong chiral medium can be equivalent to Veselago’s medium for the right circularly polarized wave \( \text{and} \) [14]. The negative \( \varepsilon_{DBF} \) and \( \mu_{DBF} \) have shown such a point. Hence the negative sign in the DBF model is not strange at all, since we realize effective double-negative with strong chirality parameter instead of simultaneously frequency resonances. For a limiting case, the chiral nilility \( \text{in} \) [11], in which \( \varepsilon \to 0 \) and \( \mu \to 0 \) while \( \kappa \neq 0 \), the parameters in DBF representation become \( \varepsilon_{DBF} \to \infty, \mu_{DBF} \to \infty \) and \( \beta = -1/(\omega \kappa) \), remaining a finite value after a simple mathematical analysis. There is no evidence that strong chirality cannot exist in this aspect.

Second, it is the effectiveness of linear models. Similar to the case that linear optical and electromagnetic models can no longer deal with very strong optical intensity and electromagnetic field, we introduce nonlinear optics to take into account the higher order terms of polarization. If the spatial dispersion is strong enough, the higher order coupling terms cannot be neglected as before [7]. People used to mistake strong chirality with strong spatial dispersion, hence adding a limitation to chirality parameter, \( \kappa < \sqrt{\mu \varepsilon} \). We believe that this is the most probable reason. However, the strong spatial dispersion is embodied in the DBF model, e.g. the value of \( \beta \), while the strong chirality is represented by the Pasteur model, e.g. the ratio of \( \kappa \) to \( \sqrt{\varepsilon \mu} \). That is to say, strong chirality does not necessarily lead to strong spatial dispersion.

Based on Eqs. (7)-(9), we have computed \( \beta \) and \( \varepsilon_{DBF} / \varepsilon \) or \( \mu_{DBF} / \mu \) versus \( \kappa / \sqrt{\varepsilon \mu} \), as shown in Figs. 1 and 2. When \( \kappa \) is very close to \( \sqrt{\mu \varepsilon} \), the value of \( \beta \) is quite large, indicating a strong spatial dispersion. Hence the singular point is the very point of traditional limitation. However, with \( \kappa \) continuously increasing, the spatial dispersion strength falls down very quickly. Therefore, if \( \kappa \) is not around \( \sqrt{\mu \varepsilon} \), e.g. \( \kappa < 0.7 \sqrt{\mu \varepsilon} \) or \( \kappa > 1.3 \sqrt{\mu \varepsilon} \), we need not take nonlinear terms into consideration at all. Hence the strong spatial dispersion and nonlinearity cannot put the upper limitation to chirality parameters either.

When \( \kappa \) is close to \( \sqrt{\mu \varepsilon} \) where the spatial dispersion is strong, we need to take higher-order terms in the DBF relations

\[
\vec{D} = \varepsilon_{DBF}(\vec{E} + \beta_1 \nabla \times \vec{E} + \beta_2 \nabla \times \nabla \times \vec{E} + \cdots), \tag{12}
\]

\[
\vec{B} = \mu_{DBF}(\vec{H} + \beta_1 \nabla \times \vec{H} + \beta_2 \nabla \times \nabla \times \vec{H} + \cdots), \tag{13}
\]

where \( \beta_n \) stands for the spatial dispersion of the \( n \)th order. We remark that the above is different
Fig. 1. The strength relationship of chirality and spatial dispersion. The point of $\kappa/\sqrt{\mu\varepsilon} = 1$ is singularity, corresponding infinite spatial dispersion coefficient $\beta$. When $\kappa/\sqrt{\mu\varepsilon} > 1$, $\beta$ becomes negative for keeping the positive rotation term coefficients with negative $\varepsilon_{DBF}$ and $\mu_{DBF}$.

Fig. 2. With chirality strength increases, $\varepsilon_{DBF}$ and $\mu_{DBF}$ reduces quickly from $\varepsilon$ and $\mu$ to $-\infty$.

from the classical nonlinear optics because it is strong spatial dispersion instead of strong field intensity. Hence it is not a power series of $\vec{E}$ and $\vec{H}$ fields. Nevertheless, the Pasteur relations should remain the form as Eqs. 5 and 6 as long as the medium is lossless and reciprocal, no matter how strong the spatial dispersion is. The only thing to be changed is the transform relation between DBF and Pasteur models, which becomes much more complicated. That is to say, though there are a lot of higher-order rotation terms, $\vec{D}$ can still be represented as a real part proportional to $\vec{E}$ and an imaginary part proportional to $\vec{H}$ with modified coefficients. $\vec{B}$ has similar representations to $\vec{D}$. The nonlinear terms contribute to the alteration of effective $\varepsilon$, $\mu$ and $\kappa$ in the Pasteur model, which might be negative, leading
to the energy problem again.

Actually, when introducing higher order terms in the DBF model, \( \varepsilon_{DBF} \) and \( \mu_{DBF} \) will be altered. Every rotation term includes real and imaginary components, related to \( \vec{E} \) and \( \vec{H} \), respectively. Comparing to the DBF model, the Pasteur model is relatively stable since its \( \varepsilon \) stands for the total proportion of \( \vec{D} \) to \( \vec{E} \). Similar conclusions are valid for \( \mu \).

Moreover, it has already been shown that any medium satisfying the Lorentz frequency-dispersive model has positive energy densities.\[15\] Using the Pasteur relations, we have

\[
\frac{dw}{dt} = \frac{\partial \vec{D}}{\partial t} \cdot \vec{E} + \frac{\partial \vec{B}}{\partial t} \cdot \vec{H} = \frac{dw'}{dt} + \text{Re} \left( i \frac{\partial (\kappa \vec{H})}{\partial t} \cdot \vec{E} - i \frac{\partial (\kappa \vec{E})}{\partial t} \cdot \vec{H} \right) = \frac{dw'}{dt} + \kappa \text{Im} \left( \frac{\partial \vec{E}}{\partial t} \cdot \vec{H} - \frac{\partial \vec{H}}{\partial t} \cdot \vec{E} \right),
\]

in which \( w' \) is the energy density in non-chiral Lorentz medium. Substituting the relation \( \vec{E} = \pm i \eta \vec{H} \) for two circularly polarized eigenwaves into above equations, the last term of Eq. (14) can be cancelled. Hence the energy density remains the same as that in the common Lorentz medium.

4. Conclusions

From Fig. 1, it is clear that enhancing spatial dispersion will not lead to strong chirality and will reach the traditional limitation point. This is why we have never succeeded in realizing strong chirality no matter how to improve the asymmetry and spatial dispersion.

Fortunately, as pointed out earlier, the strong chirality does not require strong spatial dispersion. Hence the most important difference between strong and weak chirality is that \( \kappa \) and \( \beta \) have opposite signs, which necessarily leads to negative \( \varepsilon_{DBF} \) and \( \mu_{DBF} \). Here, \( \kappa \) stands for chirality and \( \beta \) is the coefficient of the first order for spatial dispersion. Strong chirality roots from using one type of spatial dispersion to get the conjugate stereoisomer, or chirality. It is an essential condition for supporting the backward eigenwave in strong chiral medium.

In conclusion, a strong chiral medium behaves like Veselago’s medium. Under the weak spatial dispersion, the energy is always positive for chiral medium. We show that strong chirality does not equal strong spatial dispersion, which occurs only around a singular point. Even in this small region with very strong spatial dispersion, the Pasteur model is meaningful. Neither spatial dispersion nor energy will hinder chirality to be stronger, but we cannot realize strong chirality only by increasing the spatial dispersion. The necessary condition of strong chiral medium is that the chirality and spatial dispersion are of conjugated types.

We remark that strong chiral media have found wide applications in the negative refraction and supporting of backward waves, which have been discussed in details in Refs. \[14\] and \[16\]-\[21\].

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