Wave propagation in a chiral fluid: an undergraduate study

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Abstract
We study the propagation of electromagnetic waves in a chiral fluid, where the molecules are described by a simplified version of the Kuhn coupled oscillator model. The eigenmodes of Maxwell’s equations are circularly polarized waves. The application of a static magnetic field further leads to a magnetochiral term in the index of refraction of the fluid, which is independent of the wave polarization. A similar result holds when absorption is taken into account. Interference experiments and photochemical reactions have recently demonstrated the existence of the magnetochiral term. The comparison with Faraday rotation in an achiral fluid emphasizes the different symmetry properties of the two effects.

Résumé
On étudie la propagation d’ondes électromagnétiques dans un fluide chiral, dont les molécules sont décrites à l’aide d’une version simplifiée du modèle d’oscillateurs couplés de Kuhn. Les modes propres des équations de Maxwell sont des ondes polarisées circulairement. L’application d’un champ magnétique statique entraîne l’existence d’un terme magnetochiral dans l’indice de réfraction du fluide, indépendamment de la polarisation de l’onde. Un résultat semblable s’applique au cas de l’absorption. Le terme magnetochiral a été récemment mis en évidence dans des expériences d’interférences et dans des réactions photochimiques. La comparaison à la rotation Faraday dans un fluide achiral souligne les différences de symétrie des deux cas.
I. INTRODUCTION

In simple fluids, the propagation of an electromagnetic wave is usually treated as follows. Sticking to a classical description, one considers that the bound electrons of the fluid molecules are displaced from their equilibrium positions by the Lorentz force of the wave-field. This (time dependent) displacement or induced electric dipole can be modelled as a current density \( \vec{j}(\vec{r}, t) \) which is related to the electromagnetic field \( \vec{E}(\vec{r}, t) \) and \( \vec{B}(\vec{r}, t) \) through Newton’s force equation (Complications such as local fields effects will not be considered in this paper). Neglecting the magnetic contribution (the Bohr model of the hydrogen atom leads to an estimate \( |F_{\text{magn}}| \sim |\vec{r} \times \vec{B}| \sim \frac{v}{c} < 10^{-2} \)), and using Maxwell’s equations then leads to textbook expressions for the index of refraction of the fluid [1].

For chiral molecules, one has to consider the displacement of the bound electrons on a well defined geometric structure. In other words, one has to take into account the variation of the electromagnetic field down to molecular distances. The induced electric dipole moment (or current density) depends now on the field and on its spatial derivatives. This may be the reason why the propagation of electromagnetic waves in a chiral fluid is not frequently treated at an undergraduate level [2].

On the other hand, students may encounter phenomenological models which show that Maxwell’s equations in an isotropic chiral fluid have circularly polarized waves as eigenmodes [3–5]. This in turn suggests a comparison with the Faraday effect [6], which deals with the propagation of waves in a simple (achiral) fluid in the presence of a magnetic field. The comparison of these two situations raises questions about parity and time reversal transformations in a “non particle physics” context [7–10]. These symmetry considerations are not confined to theory: recent experiments [11–13] have shown that the interplay of chirality and magnetism have remarkable consequences, that have been observed in interference experiments and in photochemical reactions. Roughly speaking, the application of a magnetic field on a chiral fluid leads to a change in the index of refraction, and this new “magnetochiral” term is independent of the state of polarization of the wave. The order of magnitude of this term is rather small (see below), but its possible implications for the origin of terrestrial homochirality in biomolecules (DNA, proteins,...) are rather interesting [14].

The present paper does not claim to be original. Its merit is perhaps to bring to the attention of students some topics which are somehow scattered through the literature. Furthermore, experiments which are both “hot” and accessible to undergraduates are not so frequent.

To illustrate these considerations in a self contained way, section II will present polarization conventions and notations, as well as some definitions pertaining to chirality. Section III presents a sketchy derivation of the Faraday effect in a simple (achiral) fluid. In the framework of the elastically bound electron model, this model yields reasonable orders of magnitude, and enables one to study various aspects of the Faraday effect (dependence on the direction of propagation of light with respect to the magnetic field, reflection on a conducting mirror). The introduction of a damping term in the electron’s equation of motion can then model absorption phenomena.

In section IV, we consider a model of chiral molecules, which consists of two coupled anisotropic oscillators [15,16]. By further restricting the accessible orientations (albeit in an isotropic way), we will derive to lowest order the influence of chirality on Maxwell’s equations.
and show explicitly that the eigenmodes are circularly polarized waves. The differences with
the Faraday effect will be pointed out, in particular from the symmetry point of view.

In section V, we consider the effect of a magnetic field on a chiral fluid, along the lines
of Larmor’s theorem. This non-rigorous point of view [10] suggests the existence of a new
(symmetry allowed) term in the index of refraction, called the magnetochiral term. Its
order of magnitude is obtained through the model of section IV, and we discuss a recent
interference experiment where this term is involved. Finally, when absorption is taken
into account, the magnetochiral contribution leads to remarkable results in photochemical
reactions.

II. DEFINITIONS AND NOTATIONS

Two molecules are superposable if one can bring them into coincidence by using only
translations and rotations. A molecule (L) is chiral if it is non-superposable onto its mirror
image (D). The (L) and (D) forms of a molecule are called enantiomers of this molecule.
A fluid made of equal amounts of both enantiomers is called racemic. A fluid made only of
(L) (resp. (D)) molecules will be called a (L)-fluid (resp. a (D)-fluid) in this paper.

In agreement with many authors [16–19], we consider a wave to be right (resp. left)
circularly polarized wave if an observer looking towards the source sees at a fixed point in
space, the tip of the electric field turn clockwise (resp. anticlockwise) with time.

Physical properties pertaining to a right (resp. left) circularly polarized wave will be
denoted by the subscript $+$ (resp. $-$). A subscript L (resp. D) will be further added if one
deals with a (L)-fluid (resp. (D)-fluid). So $n_L^+(\omega)$ denotes the index of refraction of a right
circularly polarized wave of frequency $\omega$ in the (L)-fluid.

III. THE FARADAY EFFECT

A. The elastically bound electron model

We consider a monochromatic plane wave, of frequency $\omega$, propagating in a simple (achi-
ral) fluid. The direction of propagation is the $z > 0$ direction. We model the atoms as
hydrogen atoms, where the bound electrons are submitted to an elastic force $\vec{f} = -m_e\omega_0^2\vec{r}$,
where $m_e$ is the electron mass, $\omega_0$ a typical electronic frequency ($\omega_0 \sim 10^{15}$ Hz), and $\vec{r}$
is measured from the equilibrium position of the electron. Neglecting the magnetic force, we
get

$$m_e\ddot{\vec{r}} = -m_e\omega_0^2\vec{r} - e\vec{E} \tag{1}$$

If $N$ is the particle density, the induced electric dipole moment per unit volume then reads

$$\vec{P} = -Ne\vec{r} = \frac{Ne^2\vec{E}}{m_e(\omega_0^2 - \omega^2)} \tag{2}$$

leading to a polarisation current density $\vec{j} = \frac{\partial \vec{P}}{\partial t}$. Plugging these results in Maxwell’s equa-
tions [20], and denoting by $c$ the speed of light in vacuo yields
\[ \Delta \vec{E} = \frac{1}{c^2} (1 + \frac{Ne^2}{m_\varepsilon_0 (\omega_0^2 - \omega^2)}) \frac{\partial^2 \vec{E}}{\partial t^2} \] (3)

Looking for a plane wave solution \( \vec{E} = \text{Re}(\vec{E}_0 e^{i(kz - \omega t)}) \), and introducing the index of refraction \( n(\omega) \) through \( k = \frac{\omega}{c/n(\omega)} \), we get

\[ n^2(\omega) = 1 + \frac{Ne^2}{m_\varepsilon_0 (\omega_0^2 - \omega^2)} \] (4)

**B. Effect of a magnetic field and circular polarizations**

We specifically consider the propagation of a right circularly polarized wave \( (\vec{E}_+ = E_0 (\cos(kz - \omega t), \sin(kz - \omega t), 0)) \) in the presence of a static magnetic field \( \vec{B}_0 = B_0 \vec{e}_z \). The equation of motion

\[ m_\varepsilon \ddot{\vec{r}} = -m_\varepsilon \omega_0^2 \vec{r} - e(\vec{E}_+ + \dot{\vec{r}} \times \vec{B}_0) \] (5)

shows that the Lorentz force due to \( B_0 \) is antiparallel to the elastic force. Following the same steps as before gives

\[ n_+^2(\omega, B_0) = 1 + \frac{Ne^2}{m_\varepsilon_0 (\omega_0^2 - \omega^2 - \frac{eB_0}{m_\varepsilon} \omega)} \] (6)

For a left circularly polarized wave \( (\vec{E}_- = E_0 (\cos(kz - \omega t), -\sin(kz - \omega t), 0)) \), we find

\[ n_-^2(\omega, B_0) = 1 + \frac{Ne^2}{m_\varepsilon_0 (\omega_0^2 - \omega^2 + \frac{eB_0}{m_\varepsilon} \omega)} \] (7)

since in this case, the Lorentz force due to \( B_0 \) is parallel to the elastic force.

**C. Faraday rotation**

Let us now consider a linearly polarized wave propagating in a confined fluid \( (0 < z < l) \), in the presence of the magnetic field \( \vec{B}_0 = B_0 \vec{e}_z \). Since a linearly polarized wave can be decomposed into two circularly polarized waves \( (+) \) and \( (-) \) of equal amplitude but with different phase velocities \( (v_\pm = c/n_\pm(\omega, B_0)) \), elementary calculations show that the direction of vibration of the electric field rotates between \( z = 0 \) and \( z = l \) by an amount \( \alpha \), with

\[ \alpha = \frac{\omega l}{2c} \Delta n(\omega, B_0) \] (8)

where \( \Delta n(\omega, B_0) = n_+(\omega, B_0) - n_-(\omega, B_0) \). This rotation is counterclockwise for an observer looking towards the source since \( v_+ < v_- \) as shown by equations (6,7). (Note that this simple model applies only to diamagnetic materials; paramagnetic materials require a more sophisticated treatment).
Equations (6,7) also show that, as long as one deals with frequencies \( \omega \gg \omega_L = \frac{eB_0}{2m_e} \), one has

\[
n_{\pm}(\omega, B_0) \simeq n(\omega \pm \omega_L) \tag{9}
\]

up to \( O(\alpha^2) \) terms. Since \( \omega \sim \omega_0 \sim 10^{15} \) Hz and even for \( B_0 = 10 \) T, we only have \( \omega_L \sim 10^{12} \) Hz, we will consider that Larmor theorem, as expressed by equation (9), holds. This allows us to rewrite the magnetic birefringence as

\[
\Delta n(\omega, B_0) \simeq \frac{eB_0}{m_e} \frac{dn(\omega)}{d\omega} \tag{10}
\]

Typical orders of magnitude for a liquid such as CS\(_2\) are \( \Delta n(\omega, B_0) \sim 10^{-5} \) for \( B_0 = 1 \) T. The observation of the Faraday birefringence in an interference experiment is relatively easy since the Rayleigh and Michelson interferometers may detect index variations down to \( 10^{-8} \) [21].

The above expressions pertain to a wave propagating parallel to the magnetic field \( \vec{B}_0 \). For propagation in direction \( \vec{u} \) (with \( \vec{u} = \frac{\vec{k}}{k} \)), equations (9,10) read

\[
n_{\pm}(\omega, \vec{B}_0) \simeq n(\omega \pm \frac{e\vec{B}_0 \cdot \vec{u}}{2m_e}) \tag{11}
\]

and

\[
\Delta n(\omega, B_0) \simeq \frac{e(\vec{B}_0 \cdot \vec{u})}{m_e} \frac{dn(\omega)}{d\omega} \tag{12}
\]

as can be checked for a wave propagating in the \( z < 0 \) direction (\( \vec{B}_0 \cdot \vec{u} < 0 \)).

The effect of placing a perfectly conducting mirror at \( z = l \) in the original experiment (where the incident wave has \( \vec{u} = \vec{e}_z \)) can be analyzed from different points of view. The simplest one is probably described in [9]: for an observer receiving the reflected wave, the incident wave from \( z = 0 \) to \( z = l \) is equivalent, in the (symmetric w.r.t the mirror) image space, to a wave propagating in the \( z < 0 \) direction in the same magnetic field \( \vec{B}_0 = B_0 \vec{e}_z \) (this is a clear illustration of the axial character of the magnetic field \( \vec{B}_0 \), since \( \vec{B}_0 \) is perpendicular to the mirror). Taking into account the reflected wave, shows that Faraday’s rotation is doubled for this observer (the reflection on the mirror is irrelevant since it changes the sense, but not the direction of the electric field). Typical orders of magnitude for a liquid such as CS\(_2\) are \( \alpha \simeq 0.2 - 0.3 \) rd, for \( l = 10^{-2} \) m and \( \Delta n(\omega, B_0) \sim 10^{-5} \).

Finally, the inclusion of damping in equations (1,5) leads to an absorption of the wave by the fluid. Denoting by \( n_2 \) the imaginary part of the index of refraction, we clearly have \( n_{2+}(\omega, B_0) \neq n_{2-}(\omega, B_0) \). This magnetic dichroism implies an elliptical polarization (together with a Faraday rotation of the major axis of the ellipse) in the above experiments. Typical orders of magnitude are \( \frac{\Delta n_2(\omega, B_0)}{n_2(\omega)} \sim 10^{-4} - 10^{-5} \) for \( B_0 = 1 \) T.
IV. PROPAGATION IN A CHIRAL FLUID

A. The simplified Kuhn model

As previously mentioned, to take chirality into account requires a rather detailed geometric description of the fluid molecules. We consider here the simplest model of a chiral molecule [15,16], which consists of two coupled oscillators.

To fix notations, we consider a fixed trihedron \((Oxyz)\), with unit vectors \((\vec{e}_x, \vec{e}_y, \vec{e}_z)\), and a (L)-fluid with \(N\) molecules per unit volume. Each molecule has two electrons, whose equilibrium positions are called \(\vec{R}_0^1\) and \(\vec{R}_0^2\). We denote by \(d = |\vec{R}_0^1 - \vec{R}_0^2|\) the size of the molecule. We also define \(\vec{R}_{012} = \vec{R}_0^2 - \vec{R}_0^1 = d \vec{b}_{012}\) and \(\vec{R}_0 = \frac{\vec{R}_0^1 + \vec{R}_0^2}{2}\).

Due to their interaction with the propagating wave, the electrons are displaced from \(\vec{R}_0^1\) (resp. \(\vec{R}_0^2\)) by an amount \(\vec{r}_1\) (resp. \(\vec{r}_2\)).

For a fixed orientation of the molecule (i.e. for a fixed \(\vec{b}_{012}\)), the displacements of the Kuhn model are both one dimensional and coupled:

(i) the unit vector in the direction of \(\vec{r}_1\) (resp. \(\vec{r}_2\)) is denoted by \(\vec{b}_1\) (resp. \(\vec{b}_2\)). The chirality stems from the fact that the unit vectors \(\vec{b}_1, \vec{b}_2\) et \(\vec{b}_{012}\) are such that

\[
\chi = \vec{b}_{012} \cdot (\vec{b}_1 \times \vec{b}_2) = -1
\] (13)

(ii) the potential energy of the two electrons is written as

\[
E_p = \frac{1}{2} m_e \omega_0^2 (\vec{r}_1^2 + \vec{r}_2^2) + m_e \Omega_{12}^2 (\vec{b}_1 \cdot \vec{r}_1)(\vec{b}_2 \cdot \vec{r}_2)
\] (14)

where \(\omega_0\) and \(\Omega_{12}\) are electronic frequencies.

Compared to the original Kuhn model, we further add two restrictions [16]:

(iii) With respect to the fixed \((Oxyz)\) trihedron, each molecule of the fluid can only adopt the six orientations \((A_i, i = 1, 2, ..., 6)\) shown in Figure 1.

The geometrical parameters of \((A_i)\) are denoted with an index \(i\): we thus have \(\vec{b}_1^{(i)}, \vec{b}_2^{(i)}, \vec{r}_1^{(i)}, \vec{r}_2^{(i)}\), ...... The chirality is clearly the same for all orientations

\[
\chi^{(i)} = \vec{b}_{012}^{(i)} \cdot (\vec{b}_1^{(i)} \times \vec{b}_2^{(i)}) = \chi = -1.
\] (15)

(iv) We assume that the orientations \((A_i)\) are equiprobable.

These restrictions preserve the isotropy of the fluid, and make the calculations easier (more complete calculations can be found in ref [16]).

B. Equations of motion

Neglecting the magnetic contribution to the Lorentz force, we get

\[
m_e \ddot{\vec{r}}_1 = -m_e \omega_0^2 \vec{r}_1 - m_e \Omega_{12}^2 \vec{b}_1 (\vec{b}_2 \cdot \vec{r}_2) - e \vec{E} (\vec{r}_1)
\] (16)

and
\[ m_e \ddot{r}_2 = -m_e \omega_0^2 \vec{r}_2 - m_e \Omega_{12}^2 \vec{r}_2 (\vec{b}_1 \cdot \vec{r}_1) - e \vec{E}(\vec{r}_2) \] (17)

Projecting on \( \vec{b}_1 \) and \( \vec{b}_2 \), and looking for forced solutions, we obtain for orientation (A\( i \))

\[
\begin{pmatrix} \vec{r}_1 \end{pmatrix}^{(i)} = M^{-1} \begin{pmatrix} -\frac{e \vec{E}(\vec{r}_1) \cdot \vec{b}_1}{m_e} \\ \frac{e \vec{E}(\vec{r}_2) \cdot \vec{b}_2}{m_e} \end{pmatrix}^{(i)}
\] (18)

where the orientation independent matrix \( M \) is given by

\[
M = \begin{pmatrix} \omega_0^2 - \omega^2 & \Omega_{12}^2 \\ \Omega_{12}^2 & \omega_0^2 - \omega^2 \end{pmatrix}
\]

and \( M^{-1} \) is its inverse.

Let us consider for the time being a linearly polarized wave, with \( \vec{E} = \text{Re}(E(z) e^{-j\omega t})\vec{e}_x \). The resolution of the equations for each (equiprobable) orientation yields the induced electric dipole moment \( \vec{P} \) of the fluid. It will turn out that \( \vec{P} \) has components both parallel (\( \vec{P}_{//} \)) and perpendicular (\( \vec{P}_{\perp} \)) to the field \( \vec{E} \). It is easily seen that orientations (A\( 5 \), A\( 6 \)) do not contribute to \( \vec{P} \), and that orientations (A\( 3 \), A\( 4 \)) only contribute to \( \vec{P}_{//} \). On the other hand, orientations (A\( 1 \), A\( 2 \)) contribute to both components. Elementary algebra show that

\[
\vec{P}_{\perp} = \frac{N e^2}{6 m_e} \frac{\Omega_{12}^2}{(\omega_0^2 - \omega^2)^2 - \Omega_{12}^4} \left( E(z_2^{(2)}) - E(z_1^{(1)}) \right) \vec{e}_y
\] (19)

Note that the perpendicular dipole component \( \vec{P}_{\perp} \) does not vanish because of the finiteness of \( d \) (see Figure 1). To lowest order in \( d \) (or more appropriately in \( d \frac{d}{2} \)), one gets

\[
\vec{P}_{\perp} = -\frac{N e^2}{6 m_e} \frac{\Omega_{12}^2}{(\omega_0^2 - \omega^2)^2 - \Omega_{12}^4} \left( d \frac{\partial E(z)}{\partial z} \right) \vec{e}_y
\] (20)

where the gradient term is calculated at \( \vec{R}_0 \).

Gathering all contributions to \( \vec{P} \) and considering a general state of polarisation of the wave, we get

\[
\vec{P} = \alpha(\omega) \vec{E} + \gamma^L(\omega) \text{curl} \vec{E}
\] (21)

where the electric field and its derivatives are calculated at \( \vec{R}_0 \). In equation (21), one has

\[
\alpha(\omega) = \frac{2 N e^2}{3 m_e} \frac{(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 - \Omega_{12}^4}
\] (22)

and

\[
\gamma^L(\omega) = \frac{N e^2 \Omega_{12}^2 \langle \vec{R}_0 \cdot (\vec{b}_1 \times \vec{b}_2) \rangle}{6 m_e (\omega_0^2 - \omega^2)^2 - \Omega_{12}^4}
\] (23)

where we have used equation (13). Equation (23) explicitly shows that \( \gamma^L(\omega) \) is a pseudo (or axial) scalar. This feature of \( \gamma^L(\omega) \) is indeed required by equation (21), since \( \vec{P} \) is a true (or polar) vector and curl \( \vec{E} \) a pseudo (or axial) vector [23].

8
C. Rotatory power

Using equation (21) together with \( \vec{j} = \frac{\partial \vec{P}}{\partial t} \), one finds [22]

\[
\Delta \vec{E} = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \left( (1 + \frac{\alpha(\omega)}{\varepsilon_0}) \vec{E} + \frac{\gamma^L(\omega)}{\varepsilon_0} \text{curl} \ vec{E} \right)
\]

(24)

We are now back to the traditional study of Maxwell’s equations in a chiral fluid [3–5]. For circularly polarized waves in the long wavelength approximation \( (\lambda \ll 1) \), we get from equation (24)

\[
n^L_{\pm}(\omega) \simeq n(\omega) \mp \frac{\gamma^L(\omega)}{2\varepsilon_0} k_0
\]

(25)

with \( n^2(\omega) = 1 + \frac{\alpha}{\varepsilon_0} \) and \( k_0 = \frac{\omega}{c} \).

A rough order of magnitude for the natural birefringence \( (\Delta n^L(\omega) = n^L_{+}(\omega) - n^L_{-}(\omega)) \) of a chiral liquid can be obtained from \( d = 10 \ \text{Å}, \ N \sim 5 \cdot 10^{26} \ \text{m}^{-3}, \ \omega = \Omega_{12} = \frac{\omega}{2} = \pi \cdot 10^{15} \ \text{Hz} \), leading to \( \Delta n^L(\omega) \simeq 3 \cdot 10^{-5} \).

Several points can easily be checked on equations (24,25)

(i) the index of refraction is the same for a (e.g. right) circularly polarized wave propagating in the \( (L) \)-fluid, in the \( z > 0 \) and \( z < 0 \) directions.

(ii) for the \( (D) \)-fluid, one has \( n^D_{\mp}(\omega) = n^L_{\mp}(\omega) \), since \( \gamma^L(\omega) = -\gamma^D(\omega) \).

(iii) if a linearly polarized wave propagates in a \( (L) \)-fluid, confined between \( z = 0 \) and \( z = l \), the direction of vibration of the electric field will rotate by an angle \( \beta \) given by

\[
\beta = \frac{k_0 l}{2} \Delta n^L(\omega) = \frac{|\gamma^L(\omega)| k_0^2 l}{2\varepsilon_0}
\]

(26)

In our model of a \( (L) \)-fluid, the rotation is counterclockwise, and a typical order of magnitude is \( \beta \sim 1.5 \ \text{rd} \) for \( l \sim 10^{-2} \ \text{m} \).

(iv) if one places a perfectly conducting plane at \( z = l \), we again follow [9]: for an observer receiving the reflected wave, the incident wave is equivalent to a wave propagating in the (symmetric w.r.t. the mirror) image space in the \( z < 0 \) direction. In the image space, the image of the \( (L) \)-fluid is the \( (D) \)-fluid (again a clear illustration of the pseudoscalar character of chirality). The full experiment for this observer amounts to a propagation in the \( z < 0 \) direction, first through the \( (D) \)-fluid (incident wave), and then through the \( (L) \)-fluid (reflected wave), leading to a cancellation of the angle of rotation. Once again, the reflection on the mirror is irrelevant since it changes the sense, but not the direction of the electric field. This cancellation contrasts with the Faraday result and emphasizes the symmetry differences between the two cases [2,8,9].

Finally, absorption of the wave can be modelled through damping terms in equations (16,17). Detailed calculations can be found in [16]. We only stress here that the symmetry properties of the real and imaginary parts of the index of refraction are very similar. In particular, we have \( n^L_{2+}(\omega) \neq n^L_{2-}(\omega) \) (natural dichroism) and \( n^D_{2\mp}(\omega) = n^L_{2\mp}(\omega) \).

9
V. EFFECT OF A MAGNETIC FIELD: THE MAGNETOCHIRAL EFFECT

In principle, it is possible to study analytically the effect of a static magnetic field $\vec{B}_0$ on equations (16,17). It is much quicker, following [10] to apply Larmor theorem (11) to equation (25) and to write

$$n_{\pm}^L(\omega, \vec{B}_0) \sim n_{\pm}^L(\omega \pm \frac{e\vec{B}_0 \cdot \vec{u}}{2m_e})$$

for a wave propagating in direction $\vec{u}$. Expanding the right hand side of (27) leads to

$$n_{\pm}^L(\omega, \vec{B}_0) = n(\omega) \pm \frac{e(\vec{B}_0 \cdot \vec{u})}{2m_e} \frac{dn(\omega)}{d\omega} \pm \frac{\gamma^L(\omega)k_0}{2\varepsilon_0} - \frac{e}{4m_e\varepsilon_0} \frac{d\gamma^L(\omega)}{d\omega}(k_0 \cdot \vec{B}_0)$$

where $\vec{k}_0 = k_0\vec{u} = \frac{\omega}{c}\vec{u}$. Note that the Larmor frequency shift has not been done on the “$k_0$” term of equation (25). This seems plausible since the magnetic field $\vec{B}_0$ will act only on the coupled oscillators, that is on $\gamma(\omega)$; the “$k_0$” term on the other hand, comes from the operator “curl” in equation (21) which is not affected by the dynamics. We refer the reader to reference [10] for further discussions on this point.

The remarkable prediction of equation (28) is that the last term, called the magnetochiral term, does not depend on the polarization of the wave. It exists for linearly polarized or non-polarized light, and has several interesting consequences:

(i) magnetochiral birefringence: a (L)-fluid in a magnetic field and the associate (D)-fluid in the same magnetic field do not have the same index of refraction for waves propagating parallel to the field, regardless of the polarization of the waves (one may also say that a (L)-fluid in a magnetic field does not have the same index of refraction for waves propagating parallel or antiparallel to the field).

(ii) magnetochiral dichroism: the introduction of damping in the Kuhn model shows that the absorption coefficient of a (L)-fluid in a magnetic field is not the same as the absorption coefficient of a (D)-fluid, regardless of the polarization of the wave (one may also say that a (L)-fluid in a magnetic field does not absorb in the same way waves propagating parallel or antiparallel to the field).

(iii) these magnetochiral effects vanish if light propagates perpendicularly to the field.

(iv) For the mirror experiment described in section IV C, (iv), in the presence of a magnetic field $\vec{B}_0 = B_0\vec{e}_z$, the contributions of the magnetochiral term cancel. The argument is the same as for the chiral contributions.

From the Kuhn model, one may get an order of magnitude for the magnetochiral term of equation (28): setting $d = 10\,\text{A}$, $N = 5 \times 10^{26} \text{m}^{-3}$, $\omega = \Omega_{12} = \frac{\omega}{2} = \pi \times 10^{15} \text{Hz}$ and $B_0 = 10 \,\text{T}$, and using equation (23), we find a contribution to the index of refraction of order $10^{-8}$, quite close to the experimental limits [21]. One may further argue that this is precisely the order of magnitude of the terms which have been neglected in Larmor “theorem” (see equation (9)). So, one has finally to turn to experiments, which indeed have demonstrated the existence of the magnetochiral term(s).
A. Magnetochiral birefringence: an interference experiment

Imagine a Young double slit experiment with linearly polarized light, and let \( O\mathbf{z} \) be the optical axis of the set-up. After slit \( S_1 \), one adds:

- a vessel \( C_1 \) containing the (L)-fluid in a magnetic field \( \mathbf{B}_0 = B_0 \mathbf{e}_z \) followed by
- another vessel \( \overline{C}_1 \) containing the associate (D)-fluid in the opposite magnetic field \((−\mathbf{B}_0)\).

One adds after slit \( S_2 \):

- a vessel \( \overline{C}_2 \) containing the associate (D)-fluid in a field \( \mathbf{B}_0 \), followed by
- another vessel \( C_2 \) containing the (L)-fluid in the opposite magnetic field \((−\mathbf{B}_0)\).

The vessels are identical. Analyzing this experiment with equation (28) shows that the phase shift of the interference pattern caused by the enantiomers is entirely due to the magnetochiral term. All other contributions cancel, whereas the magnetochiral term is multiplied by four. The experiment has actually been done with a Michelson interferometer in a slightly modified way [12].

B. Magnetochiral dichroism: a photochemical reaction

Discriminating enantiomers is a major problem in chemistry. A classical experiment is the following (see [24,25] and references therein). One considers an initially racemic solution, that one illuminates with right circularly polarized light. The absorption of light gives rise to chemical reactions \((D \rightarrow L)\) and \((L \rightarrow D)\) [26]. In a certain range of parameters, these reactions are first order and we denote by \( k_1 \) and \( k_2 \) their respective rates. Experimentally, one finds

\[
k_1 = K I_0 n^D_{2+}(\omega) \quad \text{and} \quad k_2 = K I_0 n^L_{2+}(\omega) \quad \text{where} \quad K \text{ is a constant and } I_0 \text{ the intensity of the light.}
\]

The concentrations of the enantiomers \([L]\) and \([D]\) are given by

\[
d[L] \frac{dt}{dt} = -d[D] \frac{dt}{dt} = k_1 [D] - k_2 [L] \tag{29}
\]

At equilibrium, the solution is not racemic anymore, since one has

\[
y = \frac{[L] - [D]}{[L] + [D]} = \frac{k_1 - k_2}{k_1 + k_2} = \frac{n^D_{2+}(\omega) - n^L_{2+}(\omega)}{n^D_{2+}(\omega) + n^L_{2+}(\omega)} = \frac{n^L_{2-}(\omega) - n^L_{2+}(\omega)}{n^L_{2-}(\omega) + n^L_{2+}(\omega)} \tag{30}
\]

so that the least absorbing enantiomer is in excess.

This experiment has recently been done with natural (unpolarized) light, in the presence of a magnetic field parallel to the direction of propagation of light [13]: the magnetochiral effect implies that the absorption is not the same for the enantiomers, leading again to a non racemic solution at equilibrium. The experiment of [13], achieves a value \( y \simeq 10^{-4} \), with a magnetic field \( B_0 = 10 \ T \).
VI. CONCLUSION

We have presented in a simple way old theoretical models and recent experiments, on “chirality, light and magnetism” [27–30]. These topics can be of interest for undergraduate students, either from a physical or a (bio)chemical point of view, the more so since they also show up in other fields [31–34]. At a more advanced level, local field effects as well as quantum calculations can be introduced [35,36].

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Figure Caption

Figure 1: The allowed orientations \( \{A_i\} \) for a chiral molecule in the restricted Kuhn model. The displacement of the electrons are along \( \vec{b}_1 \) and \( \vec{b}_2 \). We consider a linearly polarized wave propagating along the \( z \) axis, with \( \vec{E} = \text{Re}(E(z) \ e^{-j\omega t})\vec{e}_x \), see text. In orientations \( \{A_1\} \) and \( \{A_2\} \), where \( \vec{b}_{12} = \vec{e}_z \), the electric field is not the same on the two electrons, leading to a non zero contribution to \( \vec{P}_\perp \), see equation (19). In orientations \( \{A_3\} \) and \( \{A_4\} \), where \( \vec{b}_{12} = \vec{e}_y \), the electric field is the same on the two electrons, leading to a contribution to \( \vec{P}_{//} \). In orientations \( \{A_5\} \) and \( \{A_6\} \), where \( \vec{b}_{12} = \vec{e}_x \), the electrons are not coupled to the electric field since \( (\vec{E} \cdot \vec{b}_1 = \vec{E} \cdot \vec{b}_2 = 0) \).
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