Reciprocity and optical chirality

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I. INTRODUCTION

Chirality (or handedness as the word stems from the greek χείρ meaning hand), refers to the lack or absence of mirror symmetry of many systems [1–3]. It is a fascinating property having important consequences in every areas of science. For example it is connected to several fundamental problems such as the apparition of life, the origin of homochirality (that is of single handedness) of many biomolecules [4], and also to the asymmetry between left and right handed fermions with respect to electroweak interaction [5]. Historically, I. Kant was one of the first eminent scholar to point out the philosophical significance of mirror operation. Already in 1783 in his celebrated “Prolegomena to any future metaphysics” he wrote

“Hence the difference between similar and equal things, which are yet not congruent (for instance, two symmetric helices), cannot be made intelligible by any concept, but only by the relation to the right and the left hands which immediately refers to intuition.” [6].

W. Thomson (Lord Kelvin), which was one of most important figure of physics at the end of the XIXth century defined more precisely chirality in the following way:

“I call any geometrical figure or group of points, chiral and say it has chirality, if its image in a plane mirror, ideally realized cannot be brought to coincide with it self.” [7]

The interest of Kelvin for chirality is not surprising. He played himself a critical role in the foundation of electromagnetism and thermodynamics, and he was well aware of the work presented in 1896 by M. Faraday on what is now known as the Faraday effect which is intimately related to optical activity and chirality [8]. In this context, it is interesting to observe that like Pasteur after him Faraday also had fruitless attempts to establish some relations between electricity, chirality and light, it was a letter from Thomson in 1845 that actually led Faraday to repeat his experiments with a magnetic field and to discover non reciprocal gyrotropy (i.e. magnetic optical rotation)!

Remarkably, this description of Kelvin provides an operational definition of chirality particularly suited to optics, as we illustrate below.

In optics indeed, since the pioneer work of Arago [9] in 1811 and Biot in 1812 [10], chirality is associated with optical activity (natural gyrotropy), which is the rotation of the plane of polarization of light upon going through a 3D chiral medium such as a quartz crystal or an aqueous solution of sugar. The first mathematical description of optical activity arisen from the work of Fresnel in 1825 [11] who interpreted phenomenologically the effect in terms of circular birefringence, that is as a difference in optical index for left and right handed circularly polarized light (respectively written LCP and RCP) passing through the medium. However, the intimate relationship between optical activity and chirality became more evident after the work of Pasteur [12, 13] in 1848 concerning the change in sign of the optical rotatory power for enantiomorphic solution of left and right handed chiral molecules of tartaric acid. In 1874, Le Bel and van’t Hoff [14, 15] related rotatory power to the unsymmetrical arrangements of substituents at a saturated atom, thus identifying the very foundation for stereochemistry. Since then, optical activity, including circular birefringence and dichroism, the so-called Cotton effect [16], that is the difference in absorption for LCP and RCP, have become very powerful probes of structural chirality in a variety of media and environments.

With the recent advent of metamaterials, that are artificially structured photonic media, a resurgence of interest concerning optical activity is observed. Current inspirations can be traced back to the pioneer work of Bose [17] who, as early as 1898, reported on the observation of rotatory power for electromagnetic microwaves propagating through a chiral artificial medium (actually left and right handed twisted jute elements). In the context of metamaterials, Lindman in 1920 [18] (see also Tinoco and Freeman in 1957 [19]) reported a similar rotatory dispersion effect through a system of copper helices in the giga-Hertz (GHz) range. Very recently, and largely due to progress in micro and
nano fabrication technics, researchers have been able to taylor compact and organized optically active metamaterials in the GHz and visible ranges. It was for instance shown, that planar chiral structures made of gammadions, i.e. equilateral crosses made of four bented arms, in metal or dielectric, can generate optical activity [24] with giant gyrotropic factors [28–30]. Important applications in opto-electronics and also for refractive devices with negative optical index for RCP and LCP, have been suggested in this context [21, 33, 37].

These studies have raised an important debate on the genuine meaning of planar chirality [21–23]. Indeed, since intuitively a two-dimensional (2D) chiral structure, which is by definition a system which can not be put into congruence with itself until left from the plane, is not expected to display any chiral optical characteristics due to the fact that simply turning the object around leads to the opposite handedness. More precisely, it was shown that since optical activity is a reciprocal property (that is obeying to the principle of reciprocity of Lorentz, see below), it necessarily implies that reversing the light path through the medium must recreate back the initial polarization state. However, since the sense of twist of a 2D chiral structure changes when looking from the second side, this polarization reversal is impossible. It would otherwise lead to the paradoxical conclusion that a left and right handed structure generates the same optical activity in contradiction with the definition, finally meaning that optical activity must vanish in strictly planar chiral systems. This behavior strongly contrasts with what is actually observed for 3D chiral objects having a helicoidal structure (like a quartz crystal [9, 12, 13], a twisted jute element [17], or a metal helix [18]), in full agreement with the principle of reciprocity of Lorentz [1, 38–41] since the sense of twist of an helix is clearly conserved when we reverse back the illumination direction. The experimental observation of optical activity in gammadion arrays forces one to conclude that such systems must present a form of hidden 3D chirality which turns fully responsible for the presence of optical rotation that rules over the dominant 2D geometrical chiral character possessed by the system.

Things could have stop here, but the understanding of 3D chirality was recently challenged in a pioneering study where it was shown that chirality has a distinct signature from optical activity when electromagnetic waves interact with a genuine 2D chiral structure and that the handedness can be recognized [42]. While the experimental demonstration was achieved in the GHz (mm) range for extended 2D structures (the so called fish-scale structures [42]), the question remained whether this could be achieved in the optical range since the optical properties of materials are not simply scalable when downsizing to the nanometer level. Theoretical suggestions were provided to overcome this difficulty by using localized plasmon modes excited at the level of the nanostructures [43]. Surface plasmons (SPs) are indeed hybrid photon/electron excitations which are naturally confined in the vicinity of a metal structure. As evanescent waves, SPs are very sensitive to local variations of the metal and dielectric environments [44]. This property was thus used to tune some 2D chiral metal structure to optical waves. Two series of experiences made in the near infrared [47] with fishscale structure and in the visible with Archimidian spirals [48, 49] confirmed the peculiarity of genuine 2D chirality at the nanoscale.

In this chapter, we will review some fundamental optical properties associated, in full generality, with chiral systems. An algebraic approach will allow us to reveal in a simple way the underlying connexions between the concepts of chirality and reciprocity from which global classes of chiral elements will be drawned. These classes will be described with the framework of Jones matrices, enabling a clear discussion on their respective optical properties. Finally, a few examples taken from our recent work will be discussed as illustrative examples of the relevant of planar chirality in the context of nanophotonics.

II. THE RECIPROCITY THEOREM AND THE PRINCIPLE OF PATH REVERSAL

A. The Lorentz reciprocity relation

In order to understand the physical meaning and implications of the different chiral matrices we will discuss we need to introduce the principle of reciprocity of Lorentz [1, 38, 39]. First, we remind that, under the validity conditions of the paraxial approximation, the properties of light going through an optical medium are fully characterized by the knowledge of the $2 \times 2$ Jones matrix

$$J := \begin{pmatrix} J_{xx} & J_{xy} \\ J_{yx} & J_{yy} \end{pmatrix},$$

which ties the incident electric field $E^{(\text{in})} = E^{(\text{in})}_x \hat{x} + E^{(\text{in})}_y \hat{y}$ to the transmitted electric field $E^{(\text{out})} = E^{(\text{out})}_x \hat{x} + E^{(\text{out})}_y \hat{y}$ (defined in the cartesian basis $x, y$ of the transverse plane). This corresponds to the transformation $E^{(\text{out})} = J \hat{E}^{(\text{in})}$, where $J$ is the operator associated with Eq. (1).

From the point of view of Jones matrices it is then possible to give a simple formulation of the reciprocity principle. Consider then a localized system illuminated by a plane wave $E^{(\text{in})}$ propagating along the $+z$ direction (see Fig.1).
FIG. 1: The two different reciprocal histories for a light beam through a medium. The initial path going from left to right is represented by the Jones matrix $J$ while the reversed path which goes from right to left corresponds to the matrix $J^{\text{rec}}$ (see Eqs. (5) and (9)).

The transmitted field (in the paraxial approximation) is given by $E^{\text{out}}(\text{in}) = \hat{J} E^{\text{in}}(\text{out})$. Same, we define also an incoming plane wave propagating along the $-z$ direction $E^{\text{in}}(\text{impinging})$ from the other side of the system and which after transmission gives the output state $E^{\text{out}}(\text{impinging}) = \hat{J}^{\text{rec.}} E^{\text{in}}(\text{impinging})$. Here by definition $\hat{J}^{\text{rec.}}$ is the reciprocal Jones operator associated with $\hat{J}$. Using Maxwell equations one can easily show that $\hat{J}^{\text{rec.}} = \hat{J}^T$ where $T$ denotes the transposition in the cartesian basis $\hat{x}, \hat{y}$. The proof reads as follow: first, from Maxwell equations one deduces \cite{1} the reciprocity theorem of Lorentz which states that if in a passive and linear environment we consider two space points $A$ and $B$ then the vector field $E(B)$ in $B$ produced by an (harmonic) point like dipole source $P(A)$ located in $A$ is linked to the field $E'(A)$ in $A$ produced by a second point like dipole $P'(B)$ located in $B$ through the formula

$$P(A) \cdot E'(A) = P'(B) \cdot E(B)$$  \hspace{1cm} (2)

(the time harmonic dependency $e^{-i\omega t}$ has been dropped everywhere).

Now, the electric field produced in $M$ by a point like dipole located in $M'$ is written $E(M) = \mathbf{G}(M, M') \cdot P(M')$ where $\mathbf{G}(M, M')$ denotes the dyadic Green function for this environment \cite{46}. Equation (2) reads thus

$$P(A) \cdot (\mathbf{G}(A, B) \cdot P'(B)) = P'(B) \cdot (\mathbf{G}(B, A) \cdot P(A))$$

or equivalently in tensorial notation

$$\sum \sum G_{ij}(A, B) - G_{ji}(B, A)P_i(A)P'_j(B) = 0 \hspace{0.5cm} (i, j = 1, 2, 3).$$

This relation is valid for every point dipoles in $A$ and $B$ and implies consequently

$$G_{ij}(A, B) = G_{ji}(B, A).$$

Actually, this relation constitutes a Maxwellian formulation of the principle of light path reversal used in optical geometry. In the next step of the proof we consider $A$ and $B$ located in $z = \pm \infty$ the fields can be then considered asymptotically as plane waves and in the paraxial approximation $G_{ij}(-\infty, +\infty)$ identifies with the Jones matrix $J_{ij}$. We immediately see that the matrix $G_{ij}(+\infty, -\infty)$ identifies with the reciprocal matrix $J^{\text{rec.}}_{ij}$. In other words, from the point of view of Jones formalism, the principle of reciprocity states

$$J^{\text{rec.}} := J^T = \begin{pmatrix} J_{xx} & J_{yx} \\ J_{xy} & J_{yy} \end{pmatrix}.$$  \hspace{1cm} (5)

In this context it is relevant to point out the similarity between the reasoning given here for establishing the reciprocity theorem and the one used in textbooks and articles \cite{1, 2, 30} for establishing the symmetry of the permittivity tensor $\epsilon_{ij}$ ($i, j = 1, 2, 3$) in solids. In particular, by taking into account spatial non-locality it is possible to obtain a version of the reciprocity theorem which reads:

$$\epsilon_{i,j}(\omega, -\mathbf{k}) = \epsilon_{j,i}(\omega, \mathbf{k})$$
where \( \mathbf{k} \) is the wavevector of the monochromatic plane wave. The analogy with Eq. (5) is complete if we choose the wave vector along the \( z \) axis and if \( i, j \) correspond to either \( x \) or \( y \). Because of these similarities many reasoning done for the Jones matrix through this chapter could be easily restated for the electric permittivity \( \epsilon \) or magnetic permeability \( \mu \) tensors.

**B. Rotation of the optical medium and reciprocity: conserving the handedness of the reference frame**

Using the previous formalism the reciprocity principle give us a univocal way to calculate the transmitted light beam propagating in the \(-z\) direction through a structure if we know the transmission Jones matrix for propagation in the \(+z\) direction. However, we must remark that this formulation is not always the most convenient since we compare the Jones matrix from a situation in which the triplet of unit vectors built by \( \hat{x}, \hat{y}, \) and the wavevector \( \mathbf{k} \) of the light wave (along \( z \) in the paraxial regime) constitute a right handed trihedra to a situation in which the same trihedra of vectors is left handed (since the sign of \( \mathbf{k} \) is opposite in the two situation). This in particular means that for an observer watching from one side \( B \) of the medium a light beam coming from the other side \( A \) the definition for LCP and RCP light as \( \hat{L}, \hat{R} = (\hat{x} \pm i\hat{y})/\sqrt{2} \) is different from the one obtained by an reciprocal observer watching from the side \( B \) a light incident from the \( A \) side, i.e., \( \hat{R}, \hat{L} = (\hat{x} \pm i\hat{y})/\sqrt{2} \). In order to remove this ambiguity the principle of reciprocity can be reformulated by considering a reference frame transformation which is a global flip of the optical medium. More precisely, comparing directly \( J^T \) to \( J \) necessitates a rotation \( R_x \) of the plane \( y-z \) by an angle \( \pi \) around \( x \). Mathematically this three dimensional transformation reads

\[
R_x = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}.
\]  

(6)

The handedness of the 3-axis coordinate system is kept unchanged after the application of the reference frame transformation \( R_x \). It also implies that the full structure of Maxwell equation is also conserved, i.e., the optical effect is rigorously equivalent to a \( \pi \)-rotation of the system around \( x \). In particular the wavevector

\[
\mathbf{k} = \begin{pmatrix} 0 \\ 0 \\ -|\mathbf{k}| \end{pmatrix}
\]

(7)

of the light going through the system transforms through \( R_x \) into

\[
\mathbf{k'} = \begin{pmatrix} 0 \\ 0 \\ +|\mathbf{k}| \end{pmatrix}.
\]

(8)

From the point of view of the 2 \( \times \) 2 Jones matrix \( R_x \) simply reduces to \( \Pi_x = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \) that is to a planar mirror symmetry with reflection axis parallel to \( x \). Through \( R_x \), the field vectors transform as \( \mathbf{E}_{Pi_x} = \hat{\Pi}_x \mathbf{E} \), so that the Jones matrix becomes \( \Pi_x \cdot J^T \cdot \Pi_x^{-1} \).

Fundamentally it is interesting to note that a symmetry operation, which is defined geometrically (and independently from any set of physical laws), manifests itself specifically according to a particular physical environment or context. From the point of view of Maxwell’s equations this symmetry operation is indeed implemented at the level of the susceptibility of the medium interaction with light that is at the level of the Jones matrices.

Actually, the reciprocal transformation defined above constitutes a rigorous and operational optical definition of the medium flipping

\[
J^{\text{flip}} := \Pi_x \cdot J^{\text{rec}} \cdot \Pi_x^{-1} \quad \text{with} \quad J^{\text{rec}} = J^T.
\]

(9)

It will be also convenient in the following to express the electric field in the left (L) and right (R) circularly polarized light basis defined by \( \hat{L}, \hat{R} = (\hat{x} \pm i\hat{y})/\sqrt{2} \). We write \( \mathcal{J} \) the Jones matrix in such basis, and we have

\[
\mathcal{J} = \begin{pmatrix} J_{ll} & J_{lr} \\ J_{rl} & J_{rr} \end{pmatrix} = U J U^{-1},
\]

(10)
The time dependence is restored after integration over the pulsation spectrum e.g., the time-reversal operation reads:

\[ \text{electric currents and charges are conveniently described by their time-Fourier transforms at the positive pulsation} \]

Maxwell’s equation be automatically satisfied by the new solutions to the variables \((\text{which immediately breaks the time-reversal symmetry.})\)

A mechanical system is time-reversal, a real system is obviously always coupled with its environment, resulting in friction and dissipation in the relation between time reversibility and reciprocity in optics. The two operators are indeed identical.

We point out that the reciprocity relations \((9)\) and \((11)\) should not be confused with the time-reversal transform.

\(J^\text{flip.}\) and \(J^\text{flip.}\) define the genuine representation of reciprocity and path reversal in a coordinate system having the same handedness as the original one (i.e. as seen from the other side of the object). This corresponds to the axes transformation \(x' = x, y' = -y, \text{ and } z' = -z\) and it implies an exchange in the role of LCP and RCP.

C. Time-reversal versus reciprocity

We point out that the reciprocity relations \((9)\) and \((11)\) should not be confused with the time-reversal transform. Time-reversal is a fundamental symmetry which dictates the invariance of physical laws between exchange of past and future. In classical mechanics, this symmetry corresponds to a system described by its position and momentum \((\mathbf{q}, \mathbf{p})\) which equations of motion are invariant through the transformation \((\mathbf{q}, \mathbf{p}, t) \rightarrow (\mathbf{q}, -\mathbf{p}, -t)\). While an isolated mechanical system is time-reversal, a real system is obviously always coupled with its environment, resulting in friction which immediately breaks the time-reversal symmetry.

In the presence of electromagnetic fields \(\mathbf{E}(x, t), \mathbf{B}(x, t)\), this time-reversal invariance is preserved if in addition to the variables \((\mathbf{q}, \mathbf{p})\) one also transforms the field into \(\mathbf{E}'(x, t) = \mathbf{E}(x, -t), \mathbf{B}'(x, t) = -\mathbf{B}'(x, -t)\). In order that Maxwell’s equation be automatically satisfied by the new solutions \(\mathbf{E}', \mathbf{B}'\) the electric current and charge distributions are changed accordingly into \(\mathbf{J}'(x, t) = -\mathbf{J}(x, -t), \rho'(x, t) = \rho(x, -t)\). In the monochromatic regime where fields, electric currents and charges are conveniently described by their time-Fourier transforms at the positive pulsation \(\omega\) the time-reversal operation reads:

\[
\begin{align*}
\mathbf{E}'(x) &= \mathbf{E}^*(x), \quad \mathbf{B}'(x) = -\mathbf{B}^*(x) \\
\mathbf{J}'(x) &= -\mathbf{J}^*(x), \quad \rho'(x) = \rho^*(x). 
\end{align*}
\]

The time dependence is restored after integration over the pulsation spectrum e.g., \(\mathbf{E}(x, t) = \int_{0}^{+\infty} \omega \mathbf{E}_\omega(x) e^{-i\omega t} + \int_{0}^{-\infty} \omega \mathbf{E}_\omega(x) e^{+i\omega t} \text{ etc...} \)

For optical situations where a modal expansion into plane-waves of vector \(k = k_x \hat{x} + k_y \hat{y}, k_z = \sqrt{(\frac{\omega}{c})^2 \varepsilon_\omega - k^2}\) is considered (\(\varepsilon_\omega\) being the complex-valued dielectric permittivity of the medium) time-reversal implies new modal components such as

\[
\begin{align*}
\varepsilon'_\omega &= \varepsilon_\omega^*, \quad k'_z = k_z^*, \quad \mathbf{E}'_{\omega, \pm}(k) = \mathbf{E}_{\omega, \mp}^*(-k), \quad \mathbf{B}'_{\omega, \pm}(k) = -\mathbf{B}_{\omega, \mp}^*(-k),
\end{align*}
\]

where by definition \(\mathbf{E}_\omega(x) = \int d^2k e^{ikx}[\mathbf{E}_{\omega, +}(k) e^{ikz} + \mathbf{E}_{\omega, -}(k) e^{-ikz}] \text{ etc...} \) In particular, if the medium is lossless, i.e., \(\text{Im} \varepsilon_\omega = 0\), the time-reversal operation dictates in the propagative sector (i.e. for \(|k| \leq \omega \sqrt{\varepsilon_\omega / c}\) a change in the sign of the wave vectors corresponding to a reversal of propagation direction for every plane-waves of the modal expansion. Going back to the Jones matrix formalism we can transform the relation \(\mathbf{E}^\text{out}\) into \(\mathbf{E}^\text{out\*}\) and \(\mathbf{J}^\text{flip\*}\) into \(\mathbf{J}^\text{flip\*}\).

Now, from the previous discussion concerning time reversibility, the complex conjugated input field \(\mathbf{E}^\text{in\*}\) at \(z = -\infty\) corresponds to the time-reversed output \(\mathbf{E}^\text{out\*}\) computed at \(z = -\infty\) whereas the complex conjugated output field \(\mathbf{E}^\text{out\*}\) at \(z = +\infty\) corresponds to the time reversed output \(\mathbf{E}^\text{in\*}\) computed at \(z = +\infty\). The Jones matrix associated with time-reversal is therefore

\[
\begin{align*}
\mathbf{J}^\text{inv} := \mathbf{J}^{-1\*} = \frac{1}{J_{xx} J_{yy} - J_{xy} J_{yx}} \begin{pmatrix}
J_{xy} J_{yy} - J_{yx} J_{yy} \\
J_{yx} J_{xx} - J_{xy} J_{xx}
end{pmatrix}
\end{align*}
\]

As it is clear from its definition, \(\mathbf{J}^\text{inv}\) is in general different from \(\mathbf{J}^\text{rec}\), exemplifying the importance of losses and dissipation in the relation between time reversibility and reciprocity in optics. The two operators are indeed identical.
if, and only if, $J$ is unitary, i.e., $J^{-1} = J^\dagger$, meaning that an optical system through which energy is conserved and which is simultaneously reciprocal will be the only optical system to be time-reversal invariant. This reveals the non-equivalence between time reversibility and reciprocity. The latter is more general: reciprocity can hold for systems in which irreversible processes take place, as a fundamental consequence of Onsager’s principle of microscopic reversibility \[50\]. In the context of planar chirality, this subtle link plays a fundamental role, as it will be discussed in section section.

### III. OPTICAL CHIRALITY

#### A. Chiral Jones Matrix

Following the operational definition of Lord Kelvin, the study of chirality demands to characterize the optical behavior of the considered system through a planar mirror symmetry $\Pi_\vartheta$. By definition, an in-plane symmetry axis making an angle $\vartheta/2$ with respect to the $x$-direction is associated with transformation matrices

$$
\Pi_\vartheta = \begin{pmatrix}
\cos \vartheta & \sin \vartheta \\
\sin \vartheta & -\cos \vartheta
\end{pmatrix},
\Pi_\vartheta^\dagger = \begin{pmatrix}
0 & e^{-i\vartheta} \\
e^{i\vartheta} & 0
\end{pmatrix},
$$

respectively written in cartesian and circular bases with $\Pi_{\vartheta=0} = \Pi_x$. Through that $\Pi_\vartheta$ symmetry operation, the Jones matrix transforms as $\hat{\mathbf{J}}_{\Pi_\vartheta} = \hat{\Pi}_\vartheta \hat{\mathbf{J}} \hat{\Pi}_\vartheta^{-1}$ in the cartesian basis and as

$$
\mathbf{J}_{\Pi} = \Pi_{\vartheta} \cdot \mathbf{J} \cdot \Pi_{\vartheta}^{-1} = \begin{pmatrix}
J_{rr} & J_{rl} e^{i2\vartheta} \\
J_{lr} e^{-i2\vartheta} & J_{ll}
\end{pmatrix}.
$$

in the circular basis.

With Kelvin’s definition, a system will be optically non-chiral if, and only if, it is invariant under $\Pi_\vartheta$, meaning that $J = J_{\Pi}$ or equivalently that the operators respectively associated with the Jones matrix and the mirror-symmetry matrix commute as $[\hat{\mathbf{J}}, \hat{\Pi}_\vartheta] = \hat{\mathbf{J}} \hat{\Pi}_\vartheta - \hat{\Pi}_\vartheta \hat{\mathbf{J}} = 0$. The invariance condition $\mathbf{J} = \mathbf{J}_{\text{mirror}}$ enforces two constraints on the Jones matrix coefficients, namely that

$$
J_{ll} = J_{rr} \quad \text{and} \quad J_{rl} = J_{lr} e^{2i\vartheta}.
$$

This implies that the Jones matrix associated with a non-chiral optical system has the following general form

$$
J_{\text{mirror}} = \begin{pmatrix}
A + B \cos \vartheta & B \sin \vartheta \\
B \sin \vartheta & A - B \cos \vartheta
\end{pmatrix}
$$

(18)

By contrapositive of conditions (17), we see that

**Theorem:** Optical chirality is possible if, and only if,

$$
J_{\Pi} \neq J_{rr} \quad \text{OR} \quad |J_{lr}| \neq |J_{rl}|
$$

OR being the logical disjunction.

This constitutes a theorem equivalent to Kelvin’s statement that an optically chiral system has no mirror symmetry, with $J \neq J_{\Pi}$ or, equivalently, with non-commuting operators respectively associated with the Jones matrix and the mirror-symmetry matrix as

$$
[J, \Pi_{\vartheta}] = J\Pi_{\vartheta} - \Pi_{\vartheta}J \neq 0 \quad \text{for any } \vartheta.
$$

(19)

Such a Jones matrix can be written in the following form:

$$
\mathbf{J} = \begin{pmatrix}
(J_{ll} + J_{rr})/2 & J_{lr} \big(J_{ll} + J_{rr}/2\big) \\
J_{rl} \big(J_{ll} + J_{rr}/2\big) & (J_{ll} - J_{rr})/2
\end{pmatrix} + \begin{pmatrix}
1 & 0 \\
0 & -1
\end{pmatrix}.
$$

(20)

Additionally for an optically chiral system, the application of a mirror symmetry $\Pi_{\vartheta}$ provides new chiral optical structures called enantiomers which are characterized by their Jones matrices $\mathbf{J}_{\text{enant.}} (\vartheta)$. By definition we have

$$
\mathbf{J}_{\text{enant.}} (\vartheta) = \Pi_{\vartheta} \cdot \mathbf{J} \cdot \Pi_{\vartheta}^{-1} \neq \mathbf{J}.
$$

(21)
In general, the lack of rotational invariance of $\mathbf{J}$ implies that these enantiomorphous matrices depend specifically on the mirror reflection $\Pi_\phi$ chosen in the definition given by Eq. (20). Thus, there are actually infinite numbers of such enantiomers.

Three important classes of chiral systems can be derived from the truth table associated with the theorem:

i) A first class satisfying $J_\| \neq J_{\|r}$ but with $|J_{lr}| = |J_{rl}|$. For reasons presented below, this class will be named the "optical activity class" or $E_{\text{o.a.}}$. This class is, until recently, the class essentially discussed in the literature.

ii) A second class corresponding to $J_{\|} = J_{\|r}$ but satisfying the constraint $|J_{lr}| \neq |J_{rl}|$. This class will be named in the following the "(genuine) planar chirality class" $E_{\text{2D}}$, for reasons also to be given further down.

iii) A third class associated with $J_{\|} \neq J_{\|r}$ and $|J_{lr}| \neq |J_{rl}|$. This is the most general class of optically chiral system coined as the "optical chirality class" and which will be described below in details.

Our point is that these chiral classes correspond to specific spatial relations of chiral systems with respect to 3D space. Such relations are fundamental to the characterization of chiral objects, which depends on the shape of the objects and the dimension of the space within which the objects are probed [51]. In optics, these relations can be unveiled through reciprocity: as light propagates in 3D space, the effect of optical path reversal through any chiral object will reveal its relation with surrounding space. Analyzing the behavior of the objects concerning reciprocity allow characterizing the relation of any chiral objects with respect to 3D space, from which a classification of the chirality type can be drawn.

\section{B. Optical activity}

One of the most illustrating example of geometrical chirality in nature is the helix. The helix is intimately linked to the most know form of optical chirality namely optical activity or natural gyrotry. For example, several natural systems like sugar molecules and quartz crystal possess a helicoidal structure and show indeed optical activity properties such as rotational power i.e. circular birefringence, or circular dichroism (i.e. a differential absorption for RCP and LCP light).

For the present purpose one of the most relevant property of helices concerns their sense of “twist”. It is indeed a basic fact that the twist orientation of an helix with its axis along Z is invariant through the rotation $R_X$: such an helix looks actually quite the same when watched by an observer in the $+Z$ or $-Z$ direction. This is mathematically rooted in the fact that the helix is a 3D object observed in a 3D space.

A particular application of the geometrical analogy is the case of an isotropic and homogenous distribution of helices which is indeed an extreme limit in which the system cannot be physically (in particular) distinguished when watched from the front or the back side. This is the case of the sugar molecules solution considered by Arago and Pasteur in their pioneer works on optical activity [9, 12]. However, it would be an oversimplification to limit optical activity to such totally invariant system since in general even an helix with its axis oriented along Z is not completely invariant through $R_X$ (although the sense of twist obviously is). Indeed, due to its finite length one will have after application of $R_X$ in general to rotate the helix by a given supplementary angle $\vartheta$ around Z in order to return to the original helix (as seen from the front side). The analogy with the helix will give us a simple way to generalize our discussion and to define a criteria for optical activity.

More precisely, in the limit of the paraxial approximation considered here the question we should ask to ourself is what must be the precise structure of the Jones matrix $\mathbf{J}$ if we impose that $J^{\text{flip}}$ (see Eq. (10)) is, up to a rotation $R_\vartheta$ by an angle $\vartheta$ around the Z axis, identical to $\mathbf{J}^\text{rl}$?

This last condition reads actually in the cartesian basis

$$ J^{\text{flip}} = \Pi_x \cdot J^{T} \cdot \Pi_x^{-1} = R_\vartheta \cdot J \cdot R_\vartheta^{-1}. \quad (22) $$

It is preferable to use the L, R basis and the previous condition becomes

$$ \mathbf{J}^{\text{T}} = \mathbf{R}_\vartheta \cdot \mathbf{J} \cdot \mathbf{R}_\vartheta^{-1}. \quad (23) $$

The rotation matrix is defined by

$$ R_\vartheta = \begin{pmatrix} \cos \vartheta & \sin \vartheta \\ -\sin \vartheta & \cos \vartheta \end{pmatrix}, \quad \mathbf{R}_\vartheta = \begin{pmatrix} e^{+i\vartheta} & 0 \\ 0 & e^{-i\vartheta} \end{pmatrix}. \quad (24) $$

From equations (9,16,22), we deduce directly that the previous condition imposes $J_{lr} e^{+i\vartheta} = J_{rl}$ (i.e., $|J_{lr}| = |J_{rl}|$). Therefore the Jones matrix takes the following form

$$ J = \begin{pmatrix} J_\| & J_{lr} e^{i\vartheta} \\ J_{rl} e^{-i\vartheta} & J_{\|r} \end{pmatrix}. \quad (25) $$
By comparing with the condition for the absence of mirror symmetry (see Eq. (103) and our theorem we see that this J matrix is chiral if and only if J_H ≠ J_r. The class of all the matrices

\[ \mathcal{J}_{\text{o.a.}} = \begin{pmatrix} J_H & J_I \\ J_I e^{i \varphi} & J_r \end{pmatrix}, \tag{26} \]

fulfilling these conditions is physically associated with the phenomenon of optical activity. This justifies the name given to E_{o.a.}. Equivalently stated this result means that Eq. (20) defines the most general Jones matrices which are i) chiral and ii) such that the optical signature of chirality is, up to a rotation \( R_\varphi \), invariant after reversal of the direction of propagation through the system. Clearly, reciprocity here dictates the rules. Importantly, Eq. (26) can also be written

\[ \mathcal{J}_{\text{o.a.}} = \begin{pmatrix} (J_H + J_r)/2 & J_I \\ J_I e^{i \varphi} & (J_H + J_r)/2 \end{pmatrix} + \frac{J_H - J_r}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \tag{27} \]

that is as the sum of a matrix \( \mathcal{J}_{\text{mirror}} \) obeying Eqs. (17,18) (i.e., having an in-plane mirror symmetry axis) and of a matrix \( \begin{pmatrix} \delta & 0 \\ 0 & -\delta \end{pmatrix} \) (with \( \delta \neq 0 \)) which actually induces the chiral behavior. In the cartesian basis we can equivalently write

\[ J_{\text{o.a.}} = \begin{pmatrix} A + B \cos \vartheta & B \sin \vartheta \\ B \sin \vartheta & A - B \cos \vartheta \end{pmatrix} + \begin{pmatrix} 0 & i\gamma \\ -i\gamma & 0 \end{pmatrix}, \tag{28} \]

with \( A = (J_H + J_r)/2, B = J_I e^{i\varphi} \) and \( \gamma = (J_r - J_H)/2 \). The presence of the antisymmetrical part is the signature of chirality and the coefficient \( \gamma \neq 0 \) is called (natural) gyromagnetic factor.

An important particular case concern Jones matrices which are invariant through a rotation by an angle \( \vartheta \) around the Z axis. Such a rotation is defined by the matrix \( R_\varphi \) with

\[ R_\varphi = \begin{pmatrix} \cos \vartheta & -\sin \vartheta \\ \sin \vartheta & \cos \vartheta \end{pmatrix}, \quad \mathcal{J}_R = \mathcal{J} R_\varphi \cdot R_\varphi^{-1} = \mathcal{J} \text{, that is:} \]

\[ \mathcal{J}_{\text{rotation axis}} = \begin{pmatrix} J_H & 0 \\ 0 & J_r \end{pmatrix}. \tag{30} \]

If \( J_H \neq J_r \) then Eq. (30) is clearly a particular case of Eq. (26) which actually describes optical activity in isotropic media, such as quartz crystals or molecular solutions, and corresponds to the circular birefringence (and dichroism) introduced by Fresnel in 1825. It is interesting to observe that this is also the matrix which is associated with the gammadions artificial structure considered in (21-24) and which have a four-fold rotational invariance around Z.

Following its definition, the Jones enantiomorphic matrix associated with \( J_{\text{o.a.}} = \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix} \) writes as \( J_{\text{o.a.}}^\text{enant.}(\vartheta) = \begin{pmatrix} B & 0 \\ 0 & A \end{pmatrix} \). Because of rotational invariance, \( J_{\text{o.a.}}^\text{enant.}(\vartheta) \) is independent of \( \vartheta \). These two enantiomorphic matrices are associated with opposite optical rotatory powers.

Consider for example the Jones matrix associated with optical activity in an isotropic medium, such as a random distribution of helices for example. From Eqs. (9,21,30) it is immediately seen that \( J_{\text{flip.}} = J \). This invariance means that an observer illuminating such a system cannot distinguish the two sides from one other. This well known property explains in particular why an optically active medium cannot be used as an optical isolator: reciprocity prohibits such a scenario. In this context we point out that nothing here forbid unitarity to hold. In the particular case of a Jones matrix represented by a rotation (see Eq. (24)) we have indeed \( J^{-1} = J^1 \). We will see in the next section that this is not the case for 2D chirality where losses are unavoidable.

It is finally useful to remark that the ensemble of all the matrices \( \mathcal{J}_{\text{o.a.}}, \) i.e., \( E_{\text{o.a.}} \), is not closed for the addition and the product of matrices (that is the sum or the product of chiral matrix belonging to \( E_{\text{o.a.}} \) is not necessarily contained in \( E_{\text{o.a.}} \)). For example by combining two enantiomers characterized by the matrices \( \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix} \) and \( \begin{pmatrix} B & 0 \\ 0 & A \end{pmatrix} \) we get

\[ \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix} + \begin{pmatrix} B & 0 \\ 0 & A \end{pmatrix} = (A + B) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \]

and, \( \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix} \cdot \begin{pmatrix} B & 0 \\ 0 & A \end{pmatrix} = (A.B) \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \tag{31} \]
which are obviously not chiral and correspond to what is called a racemic medium (that is a mixture or a juxtaposition of opposite enantiomers).

We point out that the rotation matrices considered in the present discussion involve only an axis of rotation oriented along the $z$ direction. It could be interesting to consider more general 3D rotation with an axis arbitrarily oriented. However, the case of planar chirality to be considered in the next section would thus be problematic in our classification since as stated in the introduction a planar chiral structure can be brought into congruence with its mirror image if it is lifted from the plane. The classification used here which consider explicitly as distinct the two ensembles $E_{2D}$ and $E_{o.a.}$ will appear actually very convenient since (as shown in section 3.4) any chiral matrix can be split into a first matrix belonging to $E_{2D}$ and a second matrix belonging to $E_{o.a.}$.

### C. Planar chirality

Despite its fundamental importance the previous analysis of optical activity does not exhaust the problem of chirality in optics. As we wrote in the introduction, 2D chirality characterizes, by definition, a system which cannot be put into congruence with itself until left from the plane (for a more mathematical discussion see [51, 52]). This corresponds to a different chirality class than optical activity, as it can be simply seen. If instead of a 3D helix one considers a 2D spiral contained within the $(x, y)$ plane, one has obviously a system that has a dimension lower than the dimension of its surrounding space. A flip of the structure is now possible, when it was not for the helix. As we discussed above, this discussion corresponds optically to a change of twist orientation when the light path is reversed, and clearly motivates the experimental demonstration for this second class of chiral objects which are planar.

Two geometrical examples of such planar chiral object are the Archimedean spiral which has no point symmetry and the gammadion which possesses rotational invariance. Since optically gammadion is associated with gyrotropy i.e. essentially 3D effect as discussed in the previous section we can already think that such gammadions are not genuine 2D chiral object from the optical point of view (even though it is obviously the case from basic geometrical considerations). We will here consider more in details this 2D chirality class of Jones matrices, i.e, $E_{2D}$.

The planar chirality class $E_{2D}$ was until very recently completely ignored in the literature and concerns chiral systems characterized by the conditions $J_{ll} = J_{rr}$ and a Jones matrix of the form:

$$\overline{J}_{2D} = \begin{pmatrix} J_{ll} & J_{lr} \\ J_{rl} & J_{rr} \end{pmatrix} \text{ with } |J_{lr}| \neq |J_{rl}|.$$  \hfill (32)

The condition $|J_{lr}| \neq |J_{rl}|$ actually leads to chirality. As we will show below, the equality condition on diagonal elements correspond to reciprocity.

It is also important to observe that, since Eq. (32) is different from Eqs. (17,30), the matrix $\overline{J}_{2D}$ not only has no mirror symmetries, but it has additionally no rotational invariance. This means, that $\overline{J}_{2D}$ can only be associated with chiral systems without any point symmetries, such as for example an Archimedean spiral or a fish-scale structure. A gammadion structure with its four-fold rotational invariance can not display such optical property. It should also be remarked that the fish-scale structure considered in Ref. [42] actually has a central point symmetry, i.e., a two-fold rotation axis. However from the point of view of Jones matrix such transformation is equivalent to the identity, as it is immediately seen by writing $\vartheta = \pi$ in Eq. (30), and consequently the structure of $\overline{J}$ is not constrained by such transformation.

Same as for $\overline{J}_{o.a.}$ one can define enantiomers structures by the relation

$$\overline{J}_{2D}^{\text{nant.}} = \prod_{\vartheta} \cdot \overline{J}_{2D} \cdot \prod_{\vartheta}^{-1} = \begin{pmatrix} J_{ll} & J_{lr}e^{-2i\vartheta} \\ J_{rl}e^{2i\vartheta} & J_{rr} \end{pmatrix}. \hfill (33)$$

$\overline{J}_{2D}^{\text{nant.}}$ of course belongs to $E_{2D}$.

We will now go back to the reciprocity theorem and consider the properties of planar chiral system from the point of view of path reversal. Same as for optical activity the geometrical analogy appears very convenient for characterizing the reciprocal properties of chiral planar systems. The archetype of planar chiral objects is, as we already mentioned, the Archimedean spiral with no point symmetry. Watching such a spiral from one side or the other changes obviously the sense of twist. This contrasts strongly with the case of the helix of of gammadion discussed before. This suggests the following definition: the chiral system characterized by the the Jones matrix $J$ is plan chiral if and only if Eq. (19) is satisfied and if

$$J^{\text{flip}} = \Pi_x \cdot J^T \cdot \Pi_x = R_\vartheta \cdot (\Pi_x \cdot J \cdot \Pi_x) \cdot R_\vartheta^{-1}. \hfill (34)$$
where $\Pi_x \cdot J \cdot \Pi_x$ corresponds to a particular enantiomorphic Jones matrix of $J$ (see Eq. (33)) parameterized by the angle $\vartheta$.

This condition is in the $L, R$ basis equivalent to

$$
\begin{pmatrix}
J_{ll} & J_{rl} \\
J_{lr} & J_{rr}
\end{pmatrix}
= \begin{pmatrix}
J_{rr} & J_{rl} e^{2i\vartheta} \\
J_{lr} e^{-2i\vartheta} & J_{ll}
\end{pmatrix},
$$

which admits a solution if and only if $J_{ll} = J_{rr}$ and $\vartheta = 0$ or $\pi$. Since $R_0 = -R_\pi = I$ (i.e., the identity operator) it means that the rotation plays here no role in the definition and that we could have reduced our reasoning to the condition

$$J^{flip} = \Pi_x \cdot J \cdot \Pi_x$$

i.e., $J^T = J$.

Additionally, in order to satisfy Eq. (19) we must necessarily have $|J_{lr}| \neq |J_{rl}|$. This is rigorously equivalent to the definition of the class $E_{2D}$ given above.

In the cartesian basis this means

$$J_{2D} = \begin{pmatrix}
\varepsilon_+ & \Gamma \\
\Gamma & \varepsilon_-
\end{pmatrix} = J^T_{2D}$$

with $\varepsilon_+ = J_{ll} \pm (J_{lr} + J_{rl})/2$, and $\Gamma = i(J_{lr} - J_{rl})/2$. The condition for chirality $|J_{lr}| \neq |J_{rl}|$ implies $\varepsilon_+ \neq \varepsilon_-$ and $\Gamma \neq 0$. This condition for chirality also implies a stronger restriction:

Indeed, writing the non-diagonal coefficients $J_{lr}, J_{rl}$ in the polar form $J_{lr} = ae^{i\phi}$ and $J_{rl} = be^{i\chi}$ (with $a, b$ the norms and $\phi, \chi$ the phases) we can define the ratio

$$\eta = \frac{\Gamma}{(\varepsilon_+ - \varepsilon_-)} = \frac{iJ_{lr} - J_{rl}}{J_{lr} + J_{rl}}$$

which thus becomes

$$\eta = i\frac{1 - \frac{b^2}{a^2}}{1 + \frac{b^2}{a^2}} - 2\frac{b}{a} \frac{\alpha}{1 + \frac{b^2}{a^2}} \sin (\phi - \chi).$$

The condition for chirality therefore implies:

$$\text{Imag} \left[ \frac{\Gamma}{(\varepsilon_+ - \varepsilon_-)} \right] \neq 0.$$
This implies $\frac{\alpha}{\alpha-\beta} = \alpha^*$, $\frac{\beta}{\alpha-\beta} = -\gamma^*$ and $\frac{\gamma}{\alpha-\beta} = -\beta^*$. By taking the norms of each term we deduce $|\alpha^2 - \beta\gamma| = 1$ and $|\beta| = |\gamma|$. This last equality contradicts the definition of $\mathcal{J}_{2D}$ and consequently such a planar chiral Jones matrix can not be unitary. This implies that $J_{2D}^T \neq J_{2D}^{-1}$ and that therefore $J_{2D}^{rec}$ is different from $J_{2D}^{inv}$.

In other words, a 2D chiral system provides a perfect illustration that time-reversal is necessary different from reciprocity, i.e. path reversal. Since time-reversal is a key property of fundamental physical laws at the *microscopic* level, the only solution is to assume that this breaking of time-reversal at the level of 2D chiral objects is associated with *macroscopic* irreversibility. Indeed, the imaginary part of the permittivity, for example, is connected to losses and dissipation into the environment (seen as a thermal bath) and the condition for its positivity implies a strong irreversibility in the propagation. Similarly here, 2D optical chirality means that some sources of irreversibility must be present in order to prohibit unitarity of the Jones matrix. This is an interesting example where two fundamental aspects of nature namely chirality and time irreversibility (intrinsically linked to the entropic time arrow) are intimately connected.

### D. Generalization

The most general Jones matrix $J$ characterizing a chiral medium can be written:

$$
\mathcal{J}_{\text{chiral}} = \frac{(J_{ll} + J_{rr})}{2} \begin{pmatrix} J_{lr} & (J_{ll} - J_{rr})/2 \end{pmatrix} + \frac{(J_{ll} - J_{rr})}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix},
$$

where both conditions $J_{ll} \neq J_{rr}$, $|J_{lr}| \neq |J_{ll}|$ are satisfied. This defines the optical chirality class $E_{\text{chiral}}$.

An important property is that the sum of a matrix belonging to $E_{\alpha,a}$ with a matrix of $E_{2D}$ belongs to $E_{\text{chiral}}$. To see that is obviously the case it is sufficient to remark that the sum of a matrix $\mathcal{J}_{2D}$ with a matrix $\mathcal{J}_{\alpha,a}$ can be written

$$
\mathcal{J}_{2D} + \mathcal{J}_{\alpha,a} = \mathcal{J}_{2D} + \mathcal{J}_{\text{mirror}} + \begin{pmatrix} \alpha & 0 \\ 0 & -\alpha \end{pmatrix}.
$$

However we also have

$$
\mathcal{J}_{2D} + \mathcal{J}_{\text{mirror}} = \begin{pmatrix} \alpha + \alpha' & \beta + \beta' \\ \gamma + \beta' e^{i\phi} & \alpha + \alpha' \end{pmatrix}
$$

with $|\beta| \neq |\gamma|$. This is necessary of the form $\mathcal{J}_{2D}$ since otherwise we should have $|\beta + \beta'| = |\gamma + \beta' e^{i\phi}|$ in contradiction with the condition $|\beta| \neq |\gamma|$. Reciprocally any matrices $\mathcal{J}_{2D}$ can be written as a sum $\mathcal{J}_{2D} + \mathcal{J}_{\text{mirror}}$ since from the previous result for any $\mathcal{J}_{\text{mirror}}$ the difference $\mathcal{J}_{2D} - \mathcal{J}_{\text{mirror}}$ belongs to $E_{2D}$. This means that a matrix belonging to $E_{\text{chiral}}$ can always be written as the sum of a matrix belonging to the class $E_{\alpha,a}$ with a matrix belonging to $E_{2D}$. Interestingly, the combination of a spiral and an helix leads to the geometrical shape of the screw. Finally, one can observe that $E_{\text{chiral}}$ is not close with respect to the matrix addition and product since it is already not the case for the sub classes $E_{\alpha,a}$ and $E_{2D}$.

### E. Eigenstates and chirality: time reversal versus reciprocity

In the context of reciprocity it is of practical importance to consider the backward propagation of light through the medium along the $z$ direction after path reversal by a mirror located after it (see Fig. 2). This corresponds to the following succession of events: 1) the initial state, that we write here $|in\rangle = E_x^{(in)}|x\rangle + E_y^{(in)}|y\rangle = E_L^{(in)}|L\rangle + E_R^{(in)}|R\rangle$ (instead of $E^{(in)} = E_x^{(in)}\hat{x} + E_y^{(in)}\hat{y}$ used in section [1A]), propagates through the chiral medium and we obtain afterward the new state $|2\rangle = \hat{J}|in\rangle$. 2) The reflection by the mirror induces a change in the electric field sign and also reverses the path propagation. By rotating the coordinate axes by an angle $\pi$ around the $x$ axis we preserve (as explained before) the handedness of such coordinate system as well as the positive sign of the propagation direction. This means, that in this new basis the vector $|2\rangle$ evolves as $|3\rangle = -\hat{\Pi}_x|2\rangle$. 3) The backward propagation through the medium is described by the ‘flip’ operator $\hat{J}^{\text{flip}}$ and the final state (in the new coordinate system) reads $|out\rangle = \hat{J}^{\text{flip}}|3\rangle$. We have consequently

$$
|out\rangle = -\hat{J}^{\text{flip}}\hat{\Pi}_x\hat{J}|in\rangle = -\hat{J}^{\text{flip}}J^{\text{enant}}\hat{\Pi}_x|in\rangle
$$

(47)
where by definition $\hat{J}_{\text{enant.}} = \hat{\Pi}_x \hat{J} \hat{\Pi}_x$.

In order to analyze the effect of Eq. (47) we will first study more in details the eigenstates and eigenvalues of the chiral Jones matrix discussed in this chapter. First, we consider the case of optical activity where

\[ \hat{J}_{\text{o.a.}} = \begin{pmatrix} \alpha & 0 \\ 0 & \beta \end{pmatrix} = \hat{J}_{\text{flip}}. \]

The two eigenstates are $|L\rangle$ and $|R\rangle$ corresponding to eigenvalues $\alpha$ and $\beta$ respectively. However, $\hat{J}_{\text{flip}} = \hat{J} \neq \hat{J}_{\text{enant}}$. and while the two eigenstates of $\hat{J}_{\text{enant.}}$ are still $|L\rangle$ and $|R\rangle$ the eigenvalues are now exchanged i.e. $\beta$ and $\alpha$ respectively. Therefore, a direct application of Eq. (47) on any vector $|\text{in}\rangle = a|L\rangle + b|R\rangle$ lead to

\[ |\text{out}\rangle = -\hat{J}_{\text{enant}} \hat{\Pi}_x |\text{in}\rangle = -\alpha \beta (a|L\rangle + b|R\rangle). \]

If we are only interested into the field expression in the original coordinate system we can alternatively rewrite

\[ \hat{\Pi}_x |\text{out}\rangle = -\alpha \beta (a|L\rangle + b|R\rangle) = -\alpha \beta |\text{in}\rangle. \]

This is a direct formulation of the fact that path reversal should lead us here back to the initial state $|\text{in}\rangle$ as expected. It illustrates the impact of reciprocity on propagation and show that a 3D chiral medium doesn’t act as an optical isolator. We also point out that for a loss-less ideal medium represented by a unitary rotation matrix $\hat{J}_{\text{o.a.}} = \hat{R}_\vartheta$ with eigenvalues $e^{\pm i\vartheta}$ we have exactly $\hat{\Pi}_x |\text{out}\rangle = -|\text{in}\rangle$ which, up to the minus sign coming from the mirror reflection, is a perfect illustration of time reversal and symmetry for natural optical activity.

We now consider 2D planar chirality. The eigenstates and eigenvalues of the chiral Jones matrix

\[ \hat{J}_{2\text{D}} = \begin{pmatrix} \alpha & \beta \\ \gamma & \alpha \end{pmatrix} \]

are by definition states $|\pm\rangle$ defined by $\hat{J}_{2\text{D}}|\pm\rangle = \lambda_{\pm} |\pm\rangle$. After straightforward calculations we obtain

\[ \lambda_{\pm} = \alpha \pm \sqrt{\beta \gamma}, \]

and

\[ |\pm\rangle = \frac{\sqrt{\beta} |L\rangle \pm \sqrt{\gamma} |R\rangle}{|\beta| + |\gamma|}. \]
Using similar methods we can easily find eigenstates and values of the reciprocal matrix \( J_{2D}^{\text{flip}} = J_{2D}^{\text{enant}} \) such that \( J_{2D}^{\text{flip}} |\pm\rangle_{\text{enant}}. \) The eigenvalues are the same as for \( J_{2D} \), i.e., \( \lambda_{\pm} = \lambda_{\pm} \) but the eigenvectors are now

\[
|\pm\rangle_{\text{enant}}. = \frac{\sqrt{|\lambda|} \pm \sqrt{|\beta|} \pm \sqrt{|\gamma|}}{|\beta| + |\gamma|}.
\]

Important here \( J_{\text{flip}} = J_{\text{enant}} \) and therefore by applying Eq. (47) on the initial states \( |\in\rangle = |\pm\rangle \) we get

\[
|\text{out}\rangle = - (J_{\text{flip}})^2 \hat{\Pi}_x |\in\rangle = \mp \lambda_{\pm}^2 |\pm\rangle_{\text{enant}}.
\]

where we used the relations \( \hat{\Pi}_x |L\rangle = |R\rangle, \hat{\Pi}_x |R\rangle = |L\rangle, \) and \( \hat{\Pi}_x |\pm\rangle = |\pm\rangle_{\text{enant}}. \) Like we did for optical activity we can go back to the initial coordinate system \( x, y, z \) and the final states read now

\[
\hat{\Pi}_x |\text{out}\rangle = - \lambda_{\pm}^2 |\pm\rangle.
\]

As before that again illustrates the effectiveness of the reciprocity principle. Furthermore, since the transformation is not unitary we could not obtain such a result using \( J^{\text{inv.}} \) as defined by Eq. (14).

**IV. DISCUSSION AND EXAMPLES**

As we mentioned in the introduction it is very interesting to observe that the property concerning the change of twist for genuine 2D chiral systems when watched from two different sides stirred a considerable debate in the recent year in the context of metamaterials. To understand this more in details we remind that partly boosted by practical motivations, such as the quest of negative refractive lenses or the possibility to obtain giant optical activity for applications in optoelectronics, there is currently a renewed interest in the optical activity in artificial photonic media with planar chiral structures. It was shown for instance that planar gammadion structures, which have by definition no axis of reflection but a four-fold rotational invariance, can generate optical activity with giant gyrotropic factors. Importantly, and in contrast to the usual three dimensional (3D) chiral medium (like quartz and its helicoidal structure), planar chiral structures change their observed handedness when the direction of light is reversed through the system. This paradoxically challenged Lorentz principle of reciprocity (which is known to hold for any linear non magneto-optical media) and stirred up considerable debate which came to the conclusion that optical activity cannot be a purely 2D effect and always requires a small dissymmetry between the two sides of the system.

This point becomes more clear from the previous definitions and discussion concerning chirality and reciprocity. Indeed, a gammadion being rotationally invariant its optical properties must be characterized by a Jones matrix belonging to \( E_{\text{o.a.}} \), i.e.,

\[
J_{\text{o.a.}} = \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix},
\]

with \( A \neq B \). However since geometrically the 2D gammadion change its sense of twist when watched from the other side the discussion concerning reciprocity and change of twist developed between Eqs. imposes that the Jones matrix should also belong to \( E_{2D} \), i.e.,

\[
J_{2D} = \begin{pmatrix} \alpha & \beta \\ \gamma & \alpha \end{pmatrix}
\]

with \( |\beta| \neq |\gamma| \). The only possibility for having \( J_{2D} = J_{\text{o.a.}} \) is to impose \( \gamma = \beta = 0 \) as well as \( A = B = \alpha \) and consequently to have no optical chiral signature whatsoever. This solves the paradox and shows that if gammadion generate nevertheless optical activity with giant gyrotropic factors then the system can not be purely 2D. The third dimension (such as the presence of substrate for example) is enough to break the symmetry between the two direction of transmission through the structure and there is no violation of reciprocity since the matrix has no anymore reason to satisfy the original definition of \( J_{\text{o.a.}} \) i.e. to belong to \( E_{2D} \).

We also point out that Bunn and later L. Barron already remarked that optical chirality in 3D and also in 2D (see the next section) characterizes not only the structure itself but the complete illumination protocol including the specific orientation of the incident light relatively to the structure. This idea was recently applied in the context of metamaterials by Zheludev and coworkers by demonstrating specific forms of extrinsic optical chirality in which the individual elements themselves are not chiral while the complete array of such cells is (due to specific tilts existing between the incident light) and the objects. A related scheme has also been discussed at the level of achiral plasmonic nanohole arrays.
As mentioned in the introduction the first manifestations of optical planar chirality were observed by Zheludev and coworkers [42, 43, 47] using fish-scale periodic metal strips on a dielectric substrate. The chiral structures were first realized at the mm scale for the GHz regime in 2006 [42] and soon after scaled down to the nanometer scale for studied in the near-infrared regime in 2008 [47]. Simultaneously with these last studies we realized planar chiral gratings for surface plasmons on a gold film. These structure shown in Fig. 3 (b) are Archimede spirals defined by the parametric equations

\[ x(\theta) = \pm \rho(\theta) \cos(\theta), \quad y(\theta) = \rho(\theta) \sin(\theta), \]  

with

\[ \rho(\theta) = \frac{P}{2\pi} \theta \]  

\( \theta \) varying between \( \theta_{\text{min}} = \pi \) and \( \theta_{\text{max}} = \theta_{\text{min}} + 18\pi \). The two possible signs \( \pm \) defines two enantiomers (labelled \( L \) and \( R \) on Fig. 3(b)) which are reciprocal mirror images obtained after reflection across the y-z plane \( x = 0 \). Such clockwise or anti clockwise spirals were milled on a 310 nm thick gold film using focus ion beam methods. For such Archimede’s spirals the length \( P \approx 760 \) nm plays obviously the role of a ‘radial period’ since at each increment by an angle \( \delta \theta = 2\pi \) the radius increase by an amount \( \delta \rho = P \). The structure looks like the well known “bull eye’s” circular antennas which are used to resonantly couple monochromatic light with wavelength \( \lambda \approx P \) impinging normally to the structure [56, 57]. We point out that \( P \) is actually very close to the SP wavelength \( \lambda_{\text{SP}}(\lambda_0) = 760 \) nm which corresponds to the optical wavelength \( \lambda_0 \approx 780 \) nm [48]. However, this small difference is not relevant here. To increase this similarity with an usual bull eye antenna the groove depth, and width were also selected to favor the light coupling to the grating. Importantly, we also milled a 350 diameter hole centered at the origin of the spirals (i.e. \( x = y = 0 \)) in which light can go through. Altogether the system acted as a chiral bull eye’s antenna focusing SPs at the center \( x = y = 0 \) where they interfere with the incident light before being transmitted through the hole thanks to a Fano like mechanism [58, 61]. Spectral properties of such antennas showed the typical optical resonance centered at \( \lambda \approx P \) as for their circular or elliptical cousins [45, 56, 57, 60].

Chiral optical properties of the two enantiomorphic structures were studied performing a polarization tomography...
of the light transmitted through the hole. The method described in Ref. [48, 60] is based on the experimental determination of the $4 \times 4$ Mueller matrix [62, 63]. Such a Mueller matrix $M$ characterizes the polarization transformation applied on the incident light beam with Stokes vector

$$S^{in} = \begin{pmatrix} S^0_0 \\ S^0_1 \\ S^1_1 \\ S^1_0 \end{pmatrix}.$$  \hspace{1cm} (61)

The resultant Stokes vector $S^{out} = MS^{in}$ is linked to the electric field $E = [E_x, E_y]$ transmitted through the hole. Furthermore, subsequent theoretical analysis demonstrate a precise connection between the $2 \times 2$ Jones matrix $J$ characterizing the system and the $4 \times 4$ Mueller matrix $M$. We used an home made microscope to focus and control the state of polarization (SOP) of light going through the chiral structures. In order to study experimentally the SOP conversion by our structure, we carried out a complete polarization tomography using the optical setup sketched in Fig. 3(a). A laser beam at $\lambda = 780$ nm is focused normally onto the structure by using an objective L1. The transmitted light is collected by a second objective L2 forming an Airy spot on the camera as expected since the hole behaves like a point source in an opaque gold film. In our experiments, the intensity is thus defined by taking the maximum of the Airy spot [48, 60]. The SOP of light is prepared and analyzed with half wave plates, quarter wave plates, and polarizers located before and after the objectives.

To illustrate the polarization conversion induced by the chiral object on the transmitted light we analyze on Fig. 3(c) the transformation acting on a linearly polarized input light analyzed after transmission in four orthogonal SOP wave plates, and polarizers located before and after the objective.

Lately, chiral surface plasmon modes have also been studied in relation to singular optical effects [64–67]. Indeed, near-field excitations on chiral nano structures have shown to generating orbital angular momentum (OAM) both in the near field [68–71] and the far field [72, 73]. We have just recently presented a comprehensive analysis of the OAM transfer during plasmonic in-coupling and out-coupling by chiral nanostructures at each side of a suspended metallic membrane, stressing in particular the role of a back-side structure in generating vortex beams as $e^{i\ell \varphi}$ with tunable OAM indices $\ell$.

Our device consists of a suspended thin ($h \sim 300$ nm) metallic membrane, fabricated by evaporating a metal film on a poly(vinyl formal) resin supported by a transmission electron microscopy copper grid. After evaporation, the resin is removed using a focused ion-beam (FIB), leaving a freely suspended gold membrane. Plasmonic structures are milled, in either concentric (BE) or spiral geometry on both sides of the membrane around a unique central cylindrical aperture acting as the sole transmissive element of the whole device. The general groove radial path is given in the polar $(\rho, \phi)$ basis, as $\rho_n = (n\lambda_{SP} + m\varphi\lambda_{SP}/2\pi)\hat{\rho}$, with $n$ an integer, $\lambda_{SP}$ the SP wavelength and $m$ a pitch number. Orientation conventions are chosen with respect to the light propagation direction, so that a right-handed spiral $R_m$ corresponds to $m > 0$ and a left-handed spiral $L_m$ to $m < 0$. As we summarize below, there is an close relation between the spiral pitch $m$ and the topological charge $\ell$ of the vortex generated in the near field.

Near-field generation of OAM at the front-side ($z = 0^+$) of the structured membrane can be understood by considering that each point $\rho_n$ of the groove illuminated by the incoming field is an SP point source, launching an SP wave perpendicularly to the groove. With groove widths much smaller than the illumination wavelength, the in-plane component of the generated SP field in the vicinity of the center of the structure is $E^{SP}(\rho_0, z = 0^+) \propto G \cdot E^{in}(\rho_n, z = 0^+) \cdot \hat{n}_n$, where $G = e^{ik_{SP}\rho_0}\rho_n/(|\rho_0 - \rho_n|)^{1/2}$ is the Huygens-Fresnel plasmonic propagator and $\hat{n}_n = \kappa^{-1}(d^2 \rho^2/ds^2)$ the local unit normal vector determined from the curvature $\kappa$ and the arc length $s$ of the groove. The resultant SP field is the integral of elementary point sources over the whole groove structure. As indicated by a full evaluation, we can conveniently limit the integration to radial regions $\rho_n \gg \rho_0$ where the grooves become pratically annular. This leads to $\hat{n}_n \sim -\hat{\rho}$ and therefore to a simple expression of the integrated SP field $E^{SP} = C_{in} \cdot E^{in}$,
connected to the incoming field by an in-coupling matrix

\[ C_{\text{in}}(m) \propto e^{im\phi_0} \int_0^{2\pi} d\phi e^{im\phi} e^{-ik_{SP}\rho_0 \cos \phi} \hat{\rho} \otimes \hat{\rho}, \]  

(62)

the \( \otimes \) symbol denoting a dyadic product.

Contrasting with the recent studies that have been confined to the near field, our suspended membrane opens the possibility to decouple the singular near field into the far field, with an additional structure on the back-side of the membrane connected to the front-side by the central hole. By symmetry (assuming loss-free unitarity) the out-coupling matrix is simply given as the hermitian conjugate of the in-coupling matrix, i.e.

\[ C_{\text{out}} = C_{\text{in}}^\dagger, \]  

(63)

with \( t_{ij} \) radial functions based on products of \( m_{\text{in, out}} \pm 1 \)-order Bessel functions of the first kind, as detailed in [49]. Note that we use here circular basis conventions that allow associating a positive value to an OAM induced on a right \( m > 0 \) right spiral. With respect to this chapter, these conventions are such that \(+/−\) is associated with \( \hat{L}/\hat{R} \).

This expression (63) reveals two contributions: a polarization dependent geometric phase, within the matrix, that stems from the spin-orbit coupling at the annular groove, and a factorized dynamic phase that arises due to the spiral twist of the structure [69]. In relation to what has just been described above, the structure of Eq. (63) shows that, it would belong to the general class of chiral structure, as it is not a mere 2D chiral structure, as seen from the fact that \( t_{++} \neq t_{--} \) nor does it belong to the simple optical activity class given that \( |t_{+-}| \neq |t_{-+}| \). Obviously when \( m_{\text{in}} = m_{\text{out}} = 0 \), the system is achiral and in this case, \( T \) describes a pure spin–orbit angular momentum transfer, conserving the total angular momentum [69, 74–76].

We have checked experimentally the OAM summation rules that can be drawn from this analysis and that can be gathered in a summation table. A sketch of the experiment is presented in Fig. 4. As also shown in panel (c)

FIG. 4: (a) Experimental setup: the incoming laser beam is circularly polarized using half (HWP) and quarter-wave (QWP) plates and weakly focused by a microscope objective (5×, NA=0.13). The transmitted beam is imaged by a second objective (40×, NA=0.60) and a lens tube (\( f = 200 \) mm, not shown) on a CCD camera and analyzed in the circular polarization basis by additional HWP and QWP. (b) Scanning electron microscope image of an \( R_5 \) spiral milled on the back side of a gold membrane, with \( \lambda_{SP} = 768 \) nm. (c) Intensity distribution imaged through a \( L_1 - R_5 \) structure. Labels (±, ±) correspond to the combination of circular polarization preparation and analysis. The numbers correspond to the corresponding OAM indices. Images adapted from Refs. [49].
of the figure, we have been able to generate optical beams in the far field with OAM indices up to $\ell = 8$, in perfect agreement with the expected OAM summation rules. Note that this $\ell = 8$ value is in strict relation with the chosen structures and is not a limit to our device.

Remarkably with suspended membranes, one can actually perform experimentally the path reversal operation

\[
\begin{array}{c|c|c}
+ & m_{out} - m_{in} & + \\
- & m_{out} - m_{in} + 2 & - \\
\end{array}
\]

\[
\begin{array}{c|c|c}
+ & m_{out} - m_{in} & + \\
- & m_{out} - m_{in} - 2 & - \\
\end{array}
\]

TABLE I: Far field summation rules for OAM generated through the membrane.

![Figure 5](image)

FIG. 5: Intensity distributions of the beam emerging from (a) BE-$L_1$, (b) BE-$R_1$, (c) $R_1$-BE and (d) $L_1$-BE structures, respectively. The hole diameter used in all the structures was 400 nm. Same labels as in Fig. 4. Images adapted from Refs. [49]

As displayed in Fig. 5 when comparing panels (a) and (c) and (b) and (d), the OAM measurements however turn out to be inconsistent with a simple path reversal operation. As we fully explain in [49], this discrepancy points to the pivotal role of the central aperture in the process of OAM conservation, inducing specific OAM selection rules that must be accounted for in the generation process. This discussion is beyond the scope of this summary, and we refer the reader to our manuscript for further details.

V. OUTLOOK

We have tried in this chapter to given an overview on the close relation between the chiral behavior of optical media and reciprocity. This relation has raised recently interesting issues, particularly salient in the context of metamaterial optics. As we discussed here, an algebraic approach can be useful as it allows distinguishing different classes of chiral media with respect to reciprocity. Along these lines, one is naturally led to unveil new types of chiroptical behaviors such as optical planar chirality, a totally original optical signature that contrasts with standard optical activity. Doing so, we have also stressed how reciprocity should not be confused with time-reversal invariance. A couple of recent experimental examples have been presented that nicely illustrate these intertwined relations in the realm of nanophotonics, both in terms of polarization dynamics and optical vortices.
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