Kelvin’s chirality of optical beams

Sergey Nechayev ☐,1,2,3,4 Jörg S. Eismann1,2,4,5 Rasoul Alaee ☐,3,4 Ebrahim Karimi,1,3,4 Robert W. Boyd ☐,3,4,6 and Peter Banzer1,2,4,5,*

1Max Planck Institute for the Science of Light, Staudtstrasse 2, D-91058 Erlangen, Germany
2Institute of Optics, Information and Photonics, University Erlangen-Nuremberg, Staudtstrasse 7/B2, D-91058 Erlangen, Germany
3Department of Physics, University of Ottawa, 25 Templeton St., Ottawa, Ontario K1N 6N5, Canada
4Max Planck-University of Ottawa Centre for Extreme and Quantum Photonics, 25 Templeton St., Ottawa, Ontario K1N 6N5, Canada
5Institute of Physics, University of Graz, NAWI Graz, Universitätsplatz 5, Graz, 8010 Austria
6Institute of Optics and Department of Physics and Astronomy, University of Rochester, Rochester NY 14627 USA

(Received 17 December 2020; accepted 10 February 2021; published 2 March 2021)

Geometrical chirality is a property of objects that describes a three-dimensional mirror-symmetry violation and therefore it requires a nonvanishing spatial extent. In contrary, optical chirality describes only the local handedness of electromagnetic fields and neglects the spatial geometrical structure of optical beams. In this Letter we put forward the physical significance of geometrical chirality of spatial structure of optical beams, which we term Kelvin’s chirality. Furthermore, we report on an experiment revealing the coupling of Kelvin’s chirality to optical chirality upon transmission of a focused beam through a planar medium. Our work emphasizes the importance of Kelvin’s chirality in all light-matter interaction experiments involving structured light beams with spatially inhomogeneous phase and polarization distributions.

DOI: 10.1103/PhysRevA.103.L031501

I. INTRODUCTION

Since its first definition by Lord Kelvin in 1893 [1], the term “chiral” has found its use across the fields of physics, mathematics, chemistry, and biology. Chirality describes mirror-symmetry violation—if an object cannot be superimposed with its own mirror image by means of rotations and translations, it is termed chiral [1–4]. Consequently, geometrical chirality is inherently a nonlocal three-dimensional (3D) structural property of objects that requires a nonvanishing spatial extent [5–11]. On the other hand, optical chirality is a bit less tangible. Using parity inversion transform $\hat{P}$ (a point reflection) [1–4] to define optical chirality allows electromagnetic beams to be chiral at specific points in space. The common definition of optical chirality as $C \propto \text{Im}(\mathbf{E} \cdot \mathbf{H}^*)$ [12–21], where $\mathbf{E}$ and $\mathbf{H}$ refer to the electric and magnetic field vectors, respectively, is in perfect agreement with this.

Optical chirality plays a crucial role in chiral light-matter interactions [12,16,22–24]. Tools such as circular dichroism spectroscopy are widely used for distinguishing molecular enantiomers, studying proteins’ structure, and measuring the composition of materials [25,26]. In all these scenarios, chiroptical phenomena are used to determine whether the interacting matter features geometrical chirality originating from its spatial extent.

However, polarization and phase distributions of optical beams can exhibit fascinating topological features in 3D space as well. These include knotted and linked polarization and phase singularities [27–29] and even polarization Möbius strips [30]. Some of these peculiar field topologies are structurally asymmetric upon parity inversion [31], rendering the optical beams geometrically chiral. Nonetheless, these beams do not necessarily also exhibit optical chirality $C$. Since the definition of $C$ only refers to a local arrangement of $\mathbf{E}$ and $\mathbf{H}$, it fails to describe any form of chirality originating from the spatial extent of the beams. This geometrical chirality of the spatial polarization and phase structure of optical beams (hereinafter referred to as Kelvin’s chirality or $K$) may be directly involved in chiral light-matter interaction. Indeed, the corkscrew wave fronts of linearly polarized Laguerre-Gaussian beams ($C = 0$) readily engage in chiral light-matter interactions [32–39]. Also the polarization structure of vector beams with $C = 0$ can produce circularly polarized light (CPL) upon scattering by achiral particles [40,41]. Kelvin’s chirality therefore represents an electromagnetic equivalent of geometrical chirality and refers to a certain chiral arrangement of electromagnetic fields in 3D space. It complies with the point-reflection geometrical nonlocal definition of chirality, but it cannot be described involving optical chirality $C$.

To explore the phenomenon of Kelvin’s chirality, we construct an experiment, where a normally incident polarization-structured cylindrically symmetric beam with $C = 0$ and $K \neq 0$ is focused and transmitted through a planar stratified medium. Surprisingly, we observe CPL in transmission ($C \neq 0$) with its handedness being dependent on the incident

* peter.banzer@uni-graz.at

Published by the American Physical Society under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article’s title, journal citation, and DOI. Open access publication funded by the Max Planck Society.
FIG. 1. Schematic illustration of the system. The incoming beam, propagating from left to right, is focused and collimated by two confocally aligned aplanatic microscope objectives (MOs). The sample is positioned normally to the z axis between the MOs. The red arrows depict the local polarization of the beam before and after the introduction of a relative phase of \( \pi /2 \) between radial and azimuthal polarization components.

**K.** We elucidate our results using a simple geometrical model, generalized Fresnel coefficients of the layered sample, and helicity conservation laws \([17–21,42–49]\). Our work calls for a careful evaluation of Kelvin’s chirality as a crucial component of all light-matter interaction experiments that involve light and the transmission of a relative phase of \( \pm \pi /2 \) between the MOs (Fig. 1) positioned normally to the z axis. Transmission through a stratified medium strongly depends on the polarization (\( p \), \( s \)) and angular coordinates (\( k_x \), \( k_y \)) of the focused beam. The angle of incidence of each angular component (\( k_x \), \( k_y \)) of the focused beam is defined by \( \sin^{-1}(k_p/k) \), where \( k_p = \sqrt{k_x^2 + k_y^2} \) \([50]\). Our sample is designed such that the Fresnel coefficients \( t_p(k_p) \) and \( t_s(k_p) \) for the transmitted \( p \)- and \( s \)-polarized field components at an angle of \( 30^\circ \) with respect to the surface normal acquire a \( \pi /2 \) phase difference, but have equal amplitudes \( t_p/t_s \approx \exp(i\pi /2) \) \([53]\). As a result, the transmitted field \( E_{\rho}^s \), the intensity \( S_{\rho}^s \), and the third Stokes parameters \( S_3^s \) are

\[
E_{\rho}^s(\rho) = E(\rho) [t_p(\rho) \hat{\rho} + \sigma t_s(\rho) \hat{\sigma}],
\]

\[
S_{\rho}^s(\rho) = |E(\rho)|^2 [t_p(\rho)^2 + t_s(\rho)^2],
\]

\[
S_3^s(\rho) = -2\sigma |E(\rho)|^2 [t_p(\rho) t_s(\rho)],
\]

where \( \rho = \sqrt{x^2 + y^2} = f k_p / k \) and the angle of \( 30^\circ \) is given by the condition \( \rho \approx 0.5f \) in BFP1. Equations (3) show that the transmitted beam at this angle is circularly polarized \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. This shows that Kelvin’s chirality of the incident beam \( K(\sigma) \) can perfectly couple to optical chirality \( C(\sigma) \) of the transmitted beam in a simple cylindrically symmetric scenario.

**II. THEORY**

We investigate a system of two dry aplanatic microscope objectives (MO1 and MO2) in confocal alignment, as sketched in Fig. 1, with an incident monochromatic beam propagating along the z axis. Focusing by an aplanatic MO1 converts the spatial coordinates \((x_1, y_1)\) in the back focal plane (BFP1) of MO1 to the angular coordinates of the focused beam via \( k_x = -x_1 f / k, k_y = -y_1 f / k \), where \( f \) is the focal length, \( k = 2\pi / \lambda \) is the wave number, and \( \lambda \) is the wavelength \([50]\). Collimation by MO2 induces a reverse transformation such that in BFP2 we obtain \((x_2, y_2) = (k_x f / k, k_y f / k)\). To describe the evolution of the polarization pattern, we decompose the incident beam into radial and azimuthal polarization components, which correspond to the transverse magnetic (TM or \( p \)) and transverse electric (TE or \( s \)) polarization components of the focused beam, respectively. Reference \([50]\) describes the process in detail.

Consider now a spirally polarized incident vector beam (see Fig. 1), which is an in- or \( \pi \)-out-of-phase superposition of a radially and an azimuthally polarized mode \([40,41,51,52]\). The electric field in the BFP1, which we assume to coincide with the beam’s waist position \((z = 0)\), can be written as

\[
E^{in}_{\rho} = E(\rho) [\hat{\rho} + \sigma \hat{\sigma}],
\]

where \( E(\rho) = E_0 w_0 \exp(-\rho^2 / w_0^2) \), with \( w_0 \) being the beam waist, \( E_0 \) is a constant, \( \rho \) is the radial cylindrical coordinate, \( \hat{\rho} \) and \( \hat{\phi} \) are the radial and axial unit vectors, respectively, and \( \sigma = \pm 1 \), corresponding to an in- or \( \pi \)-out-of-phase superposition. We note that parity transformation applied to the beam in Eq. (1) inverses the direction of the spiral polarization \( \hat{P}[E^p(\rho)] = E^{in}_{\rho} \) \([31,41]\). Therefore, without assigning an exact value to \( K \), we can argue that the beam in Eq. (1) possesses a nonzero Kelvin’s chirality \( K(E^{in}_{\rho}) \neq 0 \) and that a pair beams with \( \sigma = \pm 1 \) constitute a pair of chiral enantiomers, which implies that \( K(E^{in}_{\rho}) = -K(E^{in}_{\sigma}) \). At the same time, such beams feature zero optical chirality \( C \), which in the paraxial regime can be expressed as the third Stokes parameter \( S_3 \) \([12–15,17–21]\):

\[
C^m(\rho) \propto S^m_3(\rho) = 2 \text{Im} [E^{\rho}_{\rho} \cdot \hat{\rho}] [E^{\rho}_{\sigma} \cdot \hat{\sigma}] = 0. \quad (2)
\]

We introduce a planar layered structure between the MOs (Fig. 1) and perform the measurements for linear and \( \pi \)-out-of-phase superposition \( E^{in}_{\rho} \) and transverse \( \rho \)-polarization fields \( S_3^s \) with \( \rho_1, \rho_2 \). Depending on the relative angle between the LP and the q-plate, this results in a radially, azimuthally, or one of two spirally polarized doughnut-shaped beams \( S_3^s / S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. We also measured the transmitted beam at this angle is circularly polarized \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. This shows that Kelvin’s chirality of the incident beam \( K(\sigma) \) can perfectly couple to optical chirality \( C(\sigma) \) of the transmitted beam in a simple cylindrically symmetric scenario.

**III. EXPERIMENT**

Figure 2 illustrates our experimental setup \([54]\). We transmit a Gaussian beam (wavelength \( \lambda = 532 \text{ nm} \), linewidth \( \Delta \lambda_{FWHM} \approx 4 \text{ nm} \)) through a linear polarizer (LP) and a \( q \)-plate of charge \( 1/2 \) \([55,56]\). Depending on the relative angle between the LP and the \( q \)-plate, this results in a radially, azimuthally, or one of two spirally polarized doughnut-shaped beams \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. We also measured the transmitted beam at this angle is circularly polarized \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. We also measured the transmitted beam at this angle is circularly polarized \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. We also measured the transmitted beam at this angle is circularly polarized \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam. We also measured the transmitted beam at this angle is circularly polarized \( S_3^s \approx S_0^s \approx -\sigma \) with the handedness being dependent on the spatial polarization distribution of the incident beam.
First, in chiral light-matter interactions it is not exclusively the chirality of matter that couples to optical chirality \( \mathcal{C} \), but essentially the geometrical chirality of the whole experiment. For instance, structurally achiral planar metamolecules show chiroptical response at normal incidence, if the mirror symmetry is broken by the heterogeneous material composition of their constituents [57,58]. Additionally, chiroptical effects also appear if a planar structure and the \( k \) vector of an obliquely incident CPL form a geometrically chiral arrangement [59]. Similarly, linear dipoles can emit chiral light, when appropriately positioned close to an optical waveguide [60,61]. In this regard, \( \mathcal{K} \) ensures \( \mathcal{P} \)-symmetry breaking at the level of beam geometry [31].

Second, we may ask which physical conservation laws permit generation of CPL in a cylindrically symmetric system? Helicity—the projection of spin onto the propagation direction—characterizes the handedness of a beam [17–21,42–49]. Helicity is only preserved in electromagnetically dual (impedance matched) conditions, equivalent to \( t_p = t_s \) at all angles for planar systems [42,45]. Neither the stratified medium nor the glass substrate preserve helicity. Our focused beam acquires circular polarization upon transmission through the stratified medium for the same physical reason that obliquely incident plane-wave CPL at an air-glass interface acquires elliptical polarization in ellipsometry measurements, i.e., the difference of the two Fresnel coefficients.

Third, we construct the spiral beams with \( \mathcal{K} \neq 0 \) by a superposition of a radially and an azimuthally polarized beam. Neither of these beams alone possesses Kelvin’s chirality. However, the spatial polarization distributions of radial and azimuthal beams are \( \mathcal{P} \)-even and \( \mathcal{P} \)-odd, respectively, relative to a reflection plane that contains the \( z \) axis, which breaks the \( \mathcal{P} \) symmetry of their superposition [31]. Surprisingly, a \( \mathcal{P} \)-even spatial polarization distribution of the radial beam does not violate the parity odd transformation of \( \mathbf{E} \) itself, which holds at each point of space such that \( \hat{P}[\mathbf{E}(x,y,z)] = -\mathbf{E}(-x,-y,-z) \) (see also Fig. 1(c) in [41]). Moreover, we consider the geometry of spiral beams along with their direction of propagation, which renders them asymmetric under time reversal (\( \hat{T} \)) and symmetric under combined \( \mathcal{P}\hat{T} \) inversion. Previously, Barron defined these transformation properties as “false chirality” in molecular systems [2–4] and they are still a subject of active research [16]. At the same time, corkscrew wave fronts of linearly polarized Laguerre-Gaussian beams are asymmetric under \( \mathcal{P} \) and symmetric under \( \hat{T} \) reversal, respectively, rendering them “truly chiral” [31]. We envision that further classification of \( \mathcal{K} \) and transformation properties of structured beams may be necessary.

Fourth, parity asymmetric spatial distribution of an electric field can manifest itself in amplitude, phase, polarization, or a combination thereof. For instance, in our experiment \( \mathcal{K} \) is related exclusively to polarization. Our setup is insensitive to the phase distribution and any imprint of an inhomogeneous phase—including optical vortices—would not alter the distribution of \( \mathcal{C} \) in Fig. 3. Moreover, there is no general relation between \( \mathcal{K} \) and optical vortices or beams carrying orbital angular momentum (OAM) [32–39]. Similarly, there is no general relation between spin angular momentum (SAM) density and optical chirality \( \mathcal{C} \) [13,18,19,43,46,62]. In the experiment, the incident spiral beams possess \( \mathcal{K} \neq 0 \) and zero SAM, OAM and \( \mathcal{C} \), while the transmitted beams have nonzero
not acquire optical chirality since on any measurable observable. For instance, an unfocused is independent of its optical manifestations and does not rely chirality of matter, is a function of a geometrical shape, which on OAM also illustrate interaction between one we have considered. Additionally, some works focusing SAM to OAM conversion [63,64] also serve as illustrations of coupling between C and K—an reverse phenomenon to the Kelvin’s chirality and manifested as parity asymmetric spatial polarization distribution, termed here as geometrical chirality of an optical beam, similarly to SAM, OAM, C, and K [36,40,41]. However, specific cases of SAM to OAM conversion [63,64] also serve as illustrations of coupling between C and K—a reverse phenomenon to the one we have considered. Additionally, some works focusing on OAM also illustrate interaction between K and chirality of matter [32–39].

Finally, Kelvin’s chirality of optical beams, similarly to chirality of matter, is a function of a geometrical shape, which is independent of its optical manifestations and does not rely on any measurable observable. For instance, an unfocused spiral beam transmitted through the stratified sample would not acquire optical chirality since $t_p = t_r$ at normal incidence. Quantification of K is therefore as elusive a task as quantification of chirality of matter, for which many attempts have been made, but no universal measure has been established to date [5–11].

In conclusion, we presented an experiment where the geometrical chirality of an optical beam, termed here as Kelvin’s chirality and manifested as parity asymmetric spatial polarization distribution, couples to optical chirality upon transmission of a focused beam through a planar medium in a cylindrically symmetric scenario. We elucidated the underlying mechanism of chiral light-matter interaction by symmetry, material composition, helicity conservation laws, and a simple analytical model. Our results emphasize that spatially inhomogeneous phase and polarization profiles of structured light beams constitute an important degree of freedom in chiral light-matter interactions beyond optical chirality.

ACKNOWLEDGMENTS

R.A. acknowledges the support of the Alexander von Humboldt Foundation through the Feodor Lynen Return Fellowship. R.A. and R.W.B. acknowledge support through the Natural Sciences and Engineering Research Council of Canada, the Canada Research Chairs program, and the Canada First Research Excellence Fund. R.W.B. acknowledges support under US Office of Naval Research MURI award N00014-20-1-2558. S.N. and J.S.E. contributed equally to this work.

![Intensity distributions before entering the first microscope objective. The local polarization state is indicated by red arrows. Stokes parameters $S_0$, $S_1$, $S_2$, and $S_3$ measured in transmission through the stratified medium. Theoretical counterparts are plotted as insets.](image)

**FIG. 3.** Measurements for six input polarizations: (a) radial, (b) spiral $S_{1z}$, (c) azimuthal, (d) spiral $S_{1z}$, (e) linear $x$, and (f) linear $y$. (I) Intensity distributions before entering the first microscope objective. The local polarization state is indicated by red arrows. Stokes parameters $S_0$, $S_1$, $S_2$, and $S_3$ measured in transmission through the stratified medium. Theoretical counterparts are plotted as insets. $S_0$, $S_1$, and $S_2$ correspond to transmission through a glass substrate.
[31] Here parity inversion relates to a mirror reflection of the spatial structure of a beam relative to a plane that contains the propagation direction. This should not be confused with the fact that electric field is a polar vector that locally obeys $\hat{P}\left[\mathbf{E}(x, y, z)\right] = -\mathbf{E}(-x, -y, -z)$.

