A new type of light with enhanced optical chirality

Carmelo Rosales-Guzmán [1], **Karen Volke-Sepulveda** [2] and **Juan P. Torres** [1,3]

[1] *ICFO-Institut de Ciencies Fotoniques, 08860 Castelldefels, Barcelona, Spain*
[2] *Instituto de Fisica, UNAM, Apdo. Postal 20-364, 01000 Mexico D.F., Mexico*
[3] *Dep. of Signal Theory and Communications, Univ. Politecnica de Catalunya, 08034, Barcelona, Spain*

juanp.torres@icfo.es

Recently, Tang and Cohen (Science **332**, 333, 2011) have demonstrated a scheme to enhance the chiral response of molecules, which relies on the use of circularly polarized light in a standing wave configuration. Here we show a new type of light which possesses orbital angular momentum and enhanced chiral response. Unexpectedly, in the locations where the beams show enhanced optical chirality, only the longitudinal components of the electric and magnetic fields survive, showing a new way to yield optical chirality different from the usual. © 2018 Optical Society of America

OCIS codes: 260.6042, 160.1585, 260.5430

Chirality plays a crucial role in life, since most of the important molecular building blocks of life, i.e. aminoacids and sugars, come in left- or right-handed varieties. One of the most striking features of life is why most of these molecules present a specific chirality at all, since many chemical processes performed in the lab to obtain these substances give no preference for any specific form of chirality [1].

An object is chiral if it cannot be superimposed with its own mirror image. A pair of such chiral objects or systems are called enantiomorphs, or enantiomers for the special case of molecules. Enantiomers are identical in most regards, it is only in their interaction with other chiral objects that they become distinguishable [2]. Circularly polarized light (CPL) is an example of chiral object, and there is a myriad of optical phenomena, usually referred to as *optical activity*, whose origin can be reduced to the different response of molecules to right- and left-circularly polarized light.

One example of these effects is magnetic optical rotation, a rotation of the plane of polarization of a linearly polarized beam when it propagates in a chiral medium, which can be described in terms of different refractive indices for the two types of CPL. Another example is circular dichroism (CD), the different absorption rate of chiral molecules under the presence of left- and right-circularly polarized light, which translates in the transformation of a linearly polarized beam into an elliptically polarized one. Both phenomena coexist in frequency regions which present absorption.

The different rate of absorption of a chiral medium when illuminated by the two forms of CPL is generally small, which can make its detection rather demanding in some cases. Up to recently, it was thought that this response depends only on intrinsic properties of the chiral medium. But recently, a pseudoscalar quantity termed *optical chirality* (C) was introduced by Yiqiao and Cohen [3] to quantify the amount of chirality present in an arbitrarily-shaped optical field. Crucially, the inspection of this quantity shows that it should be possible to generate superchiral fields [4,5] to produce an enhancement of the amount of circular dichroism detected. This opens a whole new scenario for the detection of optical chirality, where now the shape of the optical field plays a crucial role for enhancing the detection of chirality.

Light with orbital angular momentum (OAM) is also a chiral object. Light beams with OAM show an azimuthal phase dependence in the transverse plane of the form $\sim \exp(im\varphi)$, where the index m, which can take any integer value, determines the OAM of the beam and φ is the azimuthal angle in cylindrical coordinates. In general, the spin (polarization) and orbital contributions to the total angular momentum cannot be considered separately [6]. However, in the paraxial regime, both contributions can be manipulated independently [7].

In spite of being chiral objects, all experimental efforts aimed at detecting a chiral response of molecules making use of optical beams with OAM have apparently failed [8,9]. Some theoretical investigations of the interaction of beams with OAM and molecular systems have yielded seemingly contradictory results. Within the electronic dipole approximation for diatomic molecules, and in the paraxial approximation for light beams, it was argued [10] that the internal "electronic-type" motion does not participate in any OAM change, while later on, the inclusion of electronic, rotational, vibrational and center-of-mass motion variables, seemed to demonstrate [11] that the OAM can couple to the rotational and electronic motion. Again under the paraxial approximation, it was established that OAM cannot be engaged with the chirality of a molecular system [12].

Here we will show that certain types of optical beams endowed with OAM can indeed present an enhanced local chiral response, even larger than the response that would be obtained with the usual circularly-polarized light, so in principle, they might be used to detect an enhanced circular dichroism effect. For this purpose, we make use of two basic ingredients. First, we consider a

form of light-matter interaction [2, 13] that couples the electric and magnetic fields of the optical beam to the electric $\mathbf{p}$ and magnetic $\mathbf{m}$ dipole moments of a chiral molecule, so that

$$\mathbf{p} = \mu_E \mathbf{E} + iG\mathbf{B}$$
$$\mathbf{m} = \mu_B \mathbf{B} - iG\mathbf{E}, \qquad (1)$$

where μ_E, μ_B and G are the electric, magnetic and electric-magnetic dipole polarizabilities, respectively. Even though higher-order multipoles can also contribute for light beams with general spatial shapes [14], we assume here that these contributions are sufficiently small so they can be safely neglected. Secondly, we consider Bessel light beams endowed with OAM [15]. Bessel beams are exact solutions of Maxwell equation's, and as we will see below, this departure from the usual case of the paraxial regime allows to unveil some important features not easily shown in the paraxial framework.

The time-averaged optical chirality of a CW beam of the form $\mathcal{E}(\mathbf{r}, t) = 1/2\,\mathbf{E}(\mathbf{r})\exp(ik_z z - i\omega t) + h.c.$, where ω is the angular frequency and k_z is the longitudinal component of the wave vector, writes [3]

$$C = \frac{\omega \epsilon_0}{2} \Im \left[\mathbf{E}(\mathbf{r}) \cdot \mathbf{B}^*(\mathbf{r}) \right]. \qquad (2)$$

$\Im$ stands for the imaginary part, ϵ_0 is the permittivity of vacuum, $\mathbf{E}$ and $\mathbf{B}$ are the electric and magnetic fields, respectively, and $\mathbf{r}$ is the position. For circularly polarized light with a polarization vector of the form $\hat{x} + \sigma i \hat{y}$ ($\sigma = \pm 1$), the optical chirality is $C_{CPL} = 2\sigma k U_e$, where $U_e = \epsilon_0 |\mathbf{E}|^2/4$ is the local average energy density of the field and k is the wavenumber of the light beam.

In particular, we consider a Bessel beam which propagates along the z direction of the form [15]

$$\mathbf{E}(\mathbf{r}) = E_0 \{ (\alpha\hat{x} + \beta\hat{y}) J_m(k_t\rho)\exp(im\varphi)$$
$$+ i\frac{k_t}{2k_z}\left[(\alpha + i\beta) J_{m-1}(k_t\rho)\exp[i(m-1)\varphi] \right.$$
$$\left. - (\alpha - i\beta) J_{m+1}(k_t\rho)\exp[i(m+1)\varphi]]\right\}\hat{z}, \qquad (3)$$

where J_m is the mth-order Bessel function, α and β are complex constants indicating the polarization state, (ρ, φ) are the radial and azimuthal variables in cylindrical coordinates, m is the winding number related to the OAM of the Bessel beams and k_t is the transverse component of the wave vector satisfying $k = \sqrt{k_t^2 + k_z^2}$.

The magnetic field can be computed from Eq. (3) through Maxwell's equations. Here we are interested in the field at the center of the beam ($\rho = 0$) for the cases $m = \pm 1$. Even though these are vortex beams possessing OAM, the total electric and magnetic fields at the center do not vanish; only the transverse components vanish, but the longitudinal components of the fields survive. We consider the coherent superposition, with complex weights A and B, of two OAM optical beams with indices $m = +1$ and $m = -1$, and equal linear polarizations given by $\cos\phi\hat{x} + \sin\phi\hat{y}$, i.e., $\alpha = \cos\phi$ and $\beta = \sin\phi$ in Eq. (3).

The electric and magnetic fields at the center write

$$\mathbf{E}(0) = i\frac{E_0 k_t}{2k_z}\left[A\exp(i\phi) - B\exp(-i\phi) \right]\hat{z}$$
$$\mathbf{B}(0) = -\frac{E_0 k_t}{2\omega}\left[A\exp(i\phi) + B\exp(-i\phi) \right]\hat{z} \quad (4)$$

Inserting the expressions of $\mathbf{E}(0)$ and $\mathbf{B}(0)$ into Eq. (2), the energy density U_e and the optical chirality C turns out to be

$$U_e = \frac{\epsilon_0 k_t^2 |E_0|^2}{k_z^2}\left[|A|^2 + |B|^2 - 2|A||B|\cos(2\phi - \xi) \right]$$
$$C = -\frac{\epsilon_0 k_t^2 |E_0|^2}{k_z}\left(|A|^2 - |B|^2 \right) \qquad (5)$$

where $\xi = \arg(B/A)$ is the phase difference between the complex weights A and B. Again, the optical chirality does not generally vanish at the center of the beam ($\rho = 0$), what is no longer true for all other cases with $m \neq \pm 1$.

The structure of the electric and magnetic fields which bear optical chirality is radically different from the usual form of circularly-polarized light. The fields at the center of the beam contain a single component of the field (the longitudinal component along the direction of propagation $\hat{z}$), while in the case of circularly-polarized light, there are two orthogonal components, $\hat{x}$ and $\hat{y}$, perpendicular to the direction of propagation of the beam. For instance, for $\phi = 0$, and A and B real numbers, Eq. (4) shows that there is a $\pi/2$ phase difference between the electric and magnetic fields, which is responsible for the non-zero value of the chirality. This $\pi/2$ phase difference is also typical of circular polarized light.

One of the manifestations of the presence of optical chirality is the detection of circular dichroism, which is usually quantified by means of the dissymmetry factor, $g = 2(A^+ - A^-)/(A^+ + A^-)$, where $A^{\pm}$ is the absorption rate of chiral molecules when illuminated with chiral fields of opposite chirality. For the type of interaction considered in Eq. (1), the relative dissymmetry factor writes [3] $g/g_{CPL} = C/(2kU_e)$, where g_{CPL} is the dissymmetry that would be measured with a circular polarized beam.

For a molecule located at the center of the Bessel beam, one easily obtains

$$\frac{g}{g_{CPL}} = -\frac{k_z}{k}\frac{|A|^2 - |B|^2}{|A|^2 + |B|^2 - 2|A||B|\cos(2\phi + \xi)}, \quad (6)$$

Inspection of Eq. (6) shows that when $|A| = 0$ or $|B| = 0$, the dissymmetry factor for paraxial beams ($k_z \sim k$) is nearly that of circularly polarized light, i.e. $|g/g_{CPL}| \sim 1$. And moreover, by choosing appropriate values of A and B, so that the energy density at the center of the beam is close to zero, one can obtain a large value of the dissymmetry factor, i.e. $|g/g_{CPL}| \gg 1$, the key towards observing an enhanced chiral response [3].

Fig. 1 shows the relative dissymmetry factor as a function of the polarization angle ($\phi = 0$ corresponds to polarization along $\hat{x}$, while $\phi = 90^o$ corresponds to polarization along $\hat{y}$), for some selected values of the angle
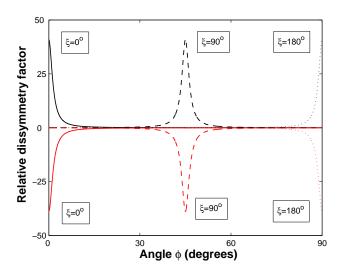
Fig. 1. (Color online) Relative dissymmetry factor g/g_{CPL} as a function of the polarization angle ϕ for two values of the ratio $r = |B|/|A|$: $r = 0.95$ (red lines) and $r = 1.05$ (black lines), and three values of the angle ξ: 0° (solid), 90° (dashed) and 180° (dotted). In all cases $k_t/k = 0.1$.

ξ and the ratio $r = |B|/|A|$. For $r = 0.95$, one obtains $g/g_{CPL} < 0$, while for $r = 1.05$, one has $g/g_{CPL} > 0$. Eq. (6) shows that an enhanced chiral response requires $|A| \sim |B|$, and $\phi = \xi/2$, so that the relative dissymmetry factor reach the maximum value of $g/g_{CPL} = (k_z/k)(|A| + |B|)/(|B| - |A|)$. The case $|A| = |B|$ would generate a null of the total electric field at the center.

Two important conclusions can be drawn from Eq. (6). Firstly, we can detect, in principle, the circular dichroism induced by a chiral molecule located at the center of a Bessel beam with winding numbers $m = \pm 1$. This is somehow unexpected, since the optical field at the center of the beam contains a single component of the electric and magnetic fields, and the transverse fields vanish. Second, the circular dichroism can even be largely enhanced when compared with the case of circularly-polarized light, similarly to the effects observed in [5] with counter-propagating circularly polarized beams. Furthermore, we want to stress that in contrast with the latter case, the superposition proposed here involves two co-propagating fields, whose centers coincide along the propagation axis, avoiding the experimental problem of locating the sample at one node of an standing wave [5].

The circular dichroism considered here could be experimentally observed by using as probe a single molecule with a fixed absorption dipole moment parallel to the beam axis [16] or a chiral solid microsphere [17], located at the center of the beam. The fluorescence of the single molecule can probe the local field intensity before and after the interaction of the light beam with the chiral medium. The probe particle can be trapped at the center of the Bessel beam by means of an auxiliary gaussian-like beam (*optical tweezer*) or by direct optical trapping with the Bessel beam itself.

In conclusion, we have unveiled a new type of light-matter interaction to obtain an enhanced chiral response. It makes use of light beams with OAM, and surprisingly, the electric and magnetic fields do not present the usual form corresponding to circularly polarized light. Notice that in the interaction of a molecule with the electric and magnetic fields, only the local fields at the position of the molecule are of interest. The approach taken here is thus in contrast to the one consider in [18,19], where the total optical chirality of the beam is considered. In that case, the optical chirality, when integrated over the whole beam, yields the global unbalance of the two spin ($\sigma = \pm 1$) angular momentum components.

References

1. P. W. Anderson, Science **177**, 393 (1972).
2. L. Barron, *Molecular light scattering and optical activity*, Cambridge University Press, 2004.
3. Y. Tang and A.E. Cohen, Phys. Rev. Lett. **104**, 163901 (2010).
4. E. Hendry et al., Nature Nanotechnology **5**, 783 (2010).
5. Y. Tang and A. E. Cohen, Science **332**, 333 (2011).
6. J. D. Jackson, *Classical electrodynamics*, John Wiley and sons, NY, 3rd edition (1999).
7. L. Allen, M. J. Padgett, and M. Babiker, Progress in Optics **39**, 291 (1999).
8. F. Araoka et al., Phys. Rev. A **71**, 055401 (2005);
9. W. Löffler et al., Phys. Rev . A **83**, 065801 (2011).
10. M. Babiker, C. R. Bennett, D. L. Andrews and L. C. Davila Romero, Phys. Rev. Lett. **89**, 143601 (2002).
11. A. Alexandrescu, D. Cojoc and E. Di Fabrizio, Phys. Rev. Lett. **96**, 243001 (2006).
12. D. L. Andrews, L. C. Davila Romero and M. Babiker, Opt. Comm. **237**, 133 (2004).
13. D. P. Craig and T. Thirunamachandran, *Molecular quantum electrodynamics*, Dover Publications Inc., 1984.
14. N. Yang and A. E. Cohen, J. Phys. Chem. **115**, 5304 (2011).
15. K. Volke-Sepulveda et al., J. Opt. B, **4**, S82 (2002).
16. L. Novotny, M. R. Beversluis, K. S. Youngworth and T. G. Brown, Phys. Rev. Lett. **86**, 5251 (2001).
17. Gabriella Cipparrone et al., Adv. Mater. **23** 7753 (2011).
18. K. Y. Bliokh and F. Nori, Phys. Rev. A **83**, 021803 (2011).
19. D. L. Andrews and M. M. Coles, Proc. SPIE **8274** 827405 (2012).

References

1. P. W. Anderson, *More is different*, Science **177**, 393 (1972).

2. L. Barron, *Molecular light scattering and optical activity*, Cambridge University Press, 2004.

3. Y. Tang et al., "Optical Chirality and its interactions with matter", Phys. Rev. Lett. **104**, 163901 (2010).

4. E. Hendry et al., "Ultrasensitive detection and characterization of biomolecules using superchiral fields", Nature Nanotechnology **5**, 783 (2010).

5. Y. Tang and A. E. Cohen, "Enhanced enantioselectivity in excitation of chiral molecules by superchiral light", Science **332**, 333 (2011).

6. J. D. Jackson, *Classical electrodynamics*, John Wiley and sons, NY, 3rd edition (1999).

7. L. Allen, M. J. Padgett, and M. Babiker,"The orbital angular momentum of light", Progress in Optics **39**, 291 (1999).

8. F. Araoka et al., "Interactions of twisted light with chiral molecules: An experimental investigation", Phys. Rev. A **71**, 055401 (2005);

9. W. Löffler et al., "Circular dichroism of cholesteric polymers and the orbital angular momentum of light", Phys. Rev . A **83**, 065801 (2011).

10. M. Babiker, C. R. Bennett, D. L. Andrews and L. C. Davila Romero, "Orbital angular momentum exchange in the interaction of twisted light with molecules", Phys. Rev. Lett. **89**, 143601 (2002).

11. A. Alexandrescu, D. Cojoc and E. Di Fabrizio, "Mechanism of angular momentum exchange between molecules and Laguerre-Gauss beams", Phys. Rev. Lett. **96**, 243001 (2006).

12. D. L. Andrews, L. C. Davila Romero and M. Babiker, "On optical vortex interaction with chiral matter", Opt. Comm. **237**, 133 (2004).

13. D. P. Craig and T. Thirunamachandran, *Molecular quantum electrodynamics*, Dover Publications Inc., 1984.

14. N. Yang and A. E. Cohen, "Local geometry of electromagnetic fields and its role in molecular multiple transtions", J. Phys. Chem. **115**, 5304 (2011).

15. K. Volke-Sepulveda et al., "Orbital angular momentum of a high-order Bessel light beam", J. Opt. B, **4**, S82 (2002).

16. L. Novotny, M. R. beversluis, K. S. Youngworth and T. G. Brown, "Longitudinal field modes probed by single molecules", Phys. Rev. Lett. **86**, 5251 (2001).

17. Gabriella Cipparrone, Alfredo Mazzulla, Alfredo Pane, Raul Josue Hernandez and Roberto Bartolino, "Chiral Self-Assembled Solid Microspheres: A Novel Multifunctional Microphotonic Device" Adv. Mater. **23** 7753 (2011).

18. K. Y. Bliokh and F. Nori, "Characterizing optical chrality", Phys. Rev. A **83**, 021803 (2011).

19. D. L. Andrews and M. M. Coles, "Optical superchirality and electromagnetic angular momentum", Proc. SPIE **8274** 827405 (2012).