Light with enhanced optical chirality

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Tang and Cohen [Phys. Rev. Lett. **104**, 163901 (2010)] recently 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 that possesses orbital angular momentum and enhanced chiral response. In the locations where the beams show enhanced optical chirality, only the longitudinal components of the electric and magnetic fields survive, which has unexpectedly shown what we believe is a new way to yield an enhanced optical chiral response.

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Chirality plays a crucial role in life because 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 systems are called 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, referred to as *optical activity*, whose origin can be traced to the different response of molecules to right- and left-CPL.

One example of these effects is 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-CPL, which translates in the transformation of a linearly polarized beam into an elliptically polarized one. Both phenomena coexist in frequency regions that 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. Until recently, it was thought that this response depends only on the intrinsic properties of the chiral medium. Recently, however, a pseudoscalar quantity, termed *optical chirality* (C), was used by Tang and Cohen [3] to quantify the amount of chiral response generated by an arbitrarily shaped optical field. Crucially, the inspection of this quantity shows that it should be possible to generate superchiral fields [4,5], which are electromagnetic fields that when they interact with chiral molecules produce an enhancement of the amount of CD 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 in enhancing the detection of chirality.

Light with orbital angular momentum (OAM) also is 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].

Theoretical investigations on the interaction of beams with OAM and molecules have yielded seemingly contradictory results. Within the electronic dipole approximation for diatomic molecules, and in the paraxial approximation, it was argued [8] 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 [9] 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 [10]. In turn, careful experiments aimed at detecting a chiral response of molecules making use of optical beams with OAM have not succeeded [11,12], apparently supporting the theoretical predictions made in [8] and [10].

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 CPL. In principle, they might be used to detect an enhanced CD 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

$$\mathbf{p} = \mu_E \mathbf{E} + iG\mathbf{B} \quad \mathbf{m} = \mu_B \mathbf{B} - iG\mathbf{E}, \tag{1}$$

where μ_E, μ_B, and G are the electric, magnetic, and electric–magnetic dipole polarizabilities, respectively.

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

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

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

$\Im$ 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 CPL with a polarization vector of the form $\hat{x} + \sigma i\hat{y}$ ($\sigma = \pm 1$), the optical chirality is $C_{\mathrm{CPL}} = 2\sigma k U_e$, where $U_e = \epsilon_0|\mathbf{E}|^2/4$ is the local average electric energy density of the field and k is the wavenumber of the light beam.

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

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

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 $\cos\phi\hat{\mathbf{x}} + \sin\phi\hat{\mathbf{y}}$, i.e., $\alpha = \cos\phi$ and $\beta = \sin\phi$ in Eq. (3).

The electric and magnetic fields at the center can be written as:

$$\begin{aligned}\mathbf{E}(0) &= i\frac{E_0 k_t}{2k_z}[A\exp(i\phi) - B\exp(-i\phi)]\hat{\mathbf{z}}\\ \mathbf{B}(0) &= -\frac{E_0 k_t}{2\omega}[A\exp(i\phi) + B\exp(-i\phi)]\hat{\mathbf{z}}.\end{aligned} \tag{4}$$

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

$$\begin{aligned}U_e &= \frac{\epsilon_0 k_t^2|E_0|^2}{16k_z^2}[|A|^2 + |B|^2 - 2|A||B|\cos(2\phi - \xi)]\\ C &= -\frac{\epsilon_0 k_t^2|E_0|^2}{8k_z}(|A|^2 - |B|^2),\end{aligned} \tag{5}$$

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

The structure of the electric and magnetic fields that bear optical chirality is radically different from the usual form of CPL. The fields at the center of the beam contain a single component of the field (along the direction of propagation $\hat{z}$), while in the case of CPL, there are two orthogonal components, $\hat{x}$ and $\hat{y}$, perpendicular to the direction of propagation. 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 nonzero value of the chirality. This $\pi/2$ phase difference is also typical of CPL.

One of the manifestations of the presence of optical chirality is the detection of CD, 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] $g/g_{\mathrm{CPL}} = C/(2kU_e)$, where g_{CPL} is the dissymmetry corresponding to a circular polarized beam.

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

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

Equation (6) shows that when $|A| = 0$ or $|B| = 0$, the dissymmetry factor for paraxial beams ($k_z \sim k$) is nearly that of CPL (i.e., $|g/g_{\mathrm{CPL}}| \sim 1$). Moreover, by choosing appropriate values of A and B, so that the electric energy density at the center of the beam is close to zero, one can enhance the dissymmetry factor (i.e., $|g/g_{\mathrm{CPL}}| \gg 1$), the key toward observing an enhanced chiral response [3].

Figure 1 shows the relative dissymmetry factor as a function of the polarization angle ($\phi = 0$ corresponds to polarization along $\hat{x}$, while $\phi = 90°$ corresponds to polarization along $\hat{y}$), for some selected values of the angle ξ and the ratio $r = |B|/|A|$. For $r = 0.95$, one obtains $g/g_{\mathrm{CPL}} < 0$, while for $r = 1.05$, one has $g/g_{\mathrm{CPL}} > 0$. Equation (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 $g/g_{\mathrm{CPL}} = (k_z/k)(|A| + |B|)/(|B| - |A|)$. 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). First, we can detect, in principle, the CD 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 CD can even be largely enhanced when compared with

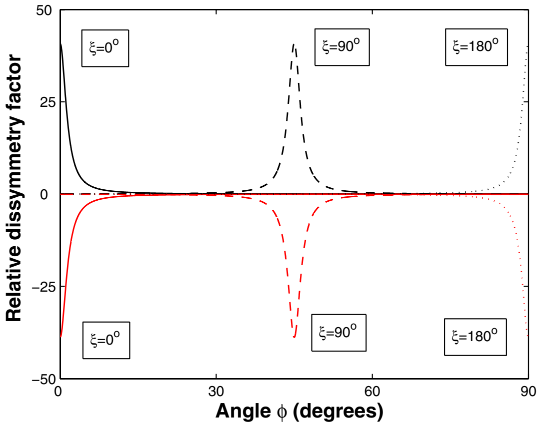

Fig. 1. (Color online) Relative dissymmetry factor as a function of the polarization angle ϕ for two values of the ratio $r = |B|/|A|$: $r = 0.95$ (red lines) and $r = 1.05$ (black lines), and three values of the angle ξ: 0° (solid), 90° (dashed) and 180° (dotted). In all cases, $k_t/k = 0.1$.

the case of CPL, which is similar to the effects observed in [5] with counter-propagating circularly polarized beams. However, the superposition proposed here involves two copropagating 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 CD considered here could be experimentally observed by using as a 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. Very recently, the longitudinal component of the electric field for the case of circularly polarized vortices with $m = \pm 1$, which plays a fundamental role in the phenomenon discussed here, has been not only detected, but used for laser ablation [18].

In conclusion, we have unveiled what we believe is 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 circular 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. When the total optical chirality of the beam, integrated over the whole beam, is also considered [19,20], it yields the global unbalance of the two spin ($\sigma = \pm 1$) angular momentum components.

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