Designing fully-structured light for chiroptical interactions

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Abstract: We derive expressions relating the forces exerted upon a chiral molecule by an optical field to gradients of the Stokes parameters of a fully-structured light beam. We show that the chiral optical force, which discriminates between molecular enantiomers, can be brought into prominence by designing a beam with a (locally) flat intensity profile but varying polarization ellipticity. Additionally, we investigate the effects of propagating these beams through a nonlinear (Kerr) medium.

1. Overview

Light beams with non-uniform intensity, phase and polarization distributions - fully-structured light (FSL) beams - have many potential applications in optical communications, high-resolution imaging, trapping and micro-particle manipulation. Here we consider FSL beams of the form

$$E(r,\varphi,z) = E_R(r,\varphi,z)\boldsymbol{e}_r + E_L(r,\varphi,z)\boldsymbol{e}_l \qquad (1)$$

where E_R and E_L consist of Laguerre-Gaussian spatial transverse modes and $\boldsymbol{e}_r$ and $\boldsymbol{e}_l$ are orthonormal circular polarization vectors [1]. The intensity and polarization distributions of these beams can be described by the Stokes parameters $S_0 = |E_R|^2 + |E_L|^2$ and $S_3 = |E_R|^2 - |E_L|^2$, respectively.

The force that light exerts on a chiral molecule can be expressed as

$$F = a\nabla w_e \pm |b|\nabla h \qquad (2)$$

where a and b pertain to the molecule, and the plus and minus signs distinguish between molecular enantiomers [2]. The first term on the right-hand side of (2) is the usual optical dipole force, which derives from gradients in the local energy density w_e, while the second term describes the chiral optical force, which derives from gradients in the local helicity density h. Typically, ($|a| \gg |b|$) and so the chiral optical force is orders of magnitude smaller than the dipole force. However, by relating w_e and h to S_0 and S_3, respectively, we can design FSL beams that locally suppress the optical dipole force while simultaneously bringing the chiral optical force to prominence, see Fig. 1.

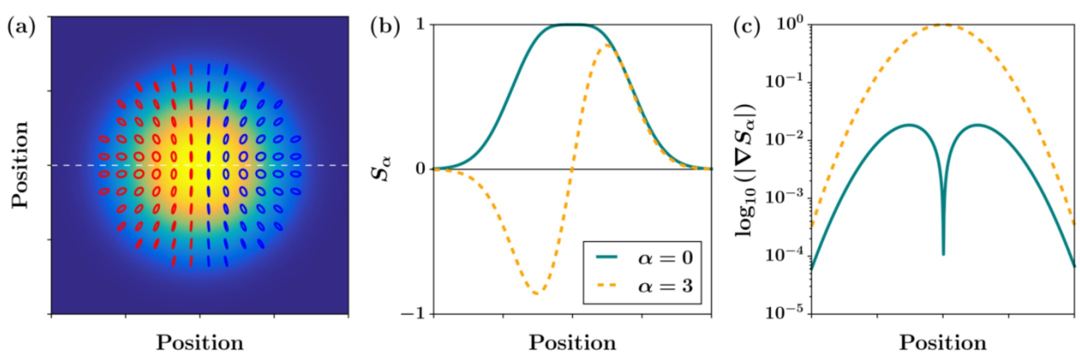

Fig. 1. (a) An intensity map of a fully-structured beam overlaid with polarization ellipses (red = left, blue = right). (b) The Stokes parameters S_0 and S_3 across the beam profile, as indicated by the white-dashed line in (a). (c) Near the center of the beam, the absolute value of the gradient of S_3 is many orders of magnitude larger than that of S_0.

It has been shown that propagation of FSL beams through a self-focusing (Kerr) nonlinear medium may counterbalance diffraction and inhibit fragmentation whilst maintaining polarization [3]. We investigate the effect that this nonlinear propagation has on the chiroptical force properties of these designed FSL beams.

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[2] R. P. Cameron, S. M. Barnett, and A. M. Yao, "Discriminatory optical force for chiral molecules," *New. J. Phys.* **16**, 013020 (2014).

[3] F. Bouchard, *et al*., "Polarization shaping for control of nonlinear propagation," *Phys. Rev. Lett*. **117**, 233903 (2016).