Structured Light Chirality: Past, Present, and Future

Kayn A. Forbes

School of Chemistry, University of East Anglia, Norwich, Norfolk, NR4 7TJ, United Kingdom

k.forbes@uea.ac.uk

Abstract: In this talk I will give a survey of the past, present, and future of the rapidly emerging field of structured light chirality, ranging from the underlying general mechanisms to state-of-the-art experiments.

1. Structured light chirality: past

For nearly 200 years, light's chirality has been associated with states of circular polarization. Electromagnetic field vectors trace out helical structures on propagation (Fig. 1) with either a left-handed $\sigma = 1$ or right-handed $\sigma = -1$ twist. This optical chirality can engage differentially with material chirality in light-matter interactions through optical activity, leading to chiral optical forces and spectroscopies. The latter are an invaluable tool in determining the absolute configurations and dynamics of drugs and biomolecules, such as proteins and viruses.

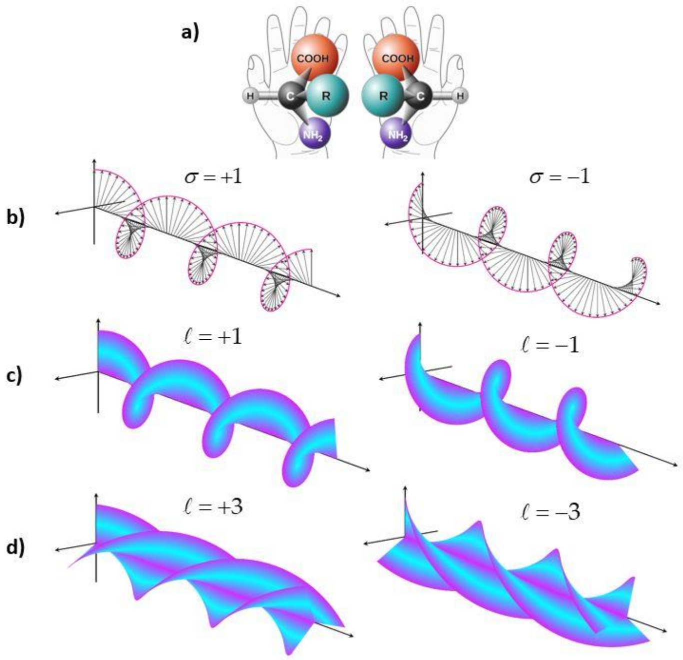

Figure 1: a) material chirality; b) circular polarization) c) and d) Optical vortex helical wavefronts.

Our ability to readily tailor laser beam modes has led to the field of structured light [1]. Structured light can possess chiral structures distinct from circular polarization. An important example are optical vortex modes which propagate with the azimuthal phase $\exp(i\ell\phi)$ leading to chiral helical wavefronts (Fig. 1). The magnitude of the topological charge $\ell \in \mathbb{Z}$ determines the number of intertwined helical wavefronts, while the sign determines their handedness: $\ell > 0$ are left-handed; $\ell < 0$ are right-handed. In the early 2000s researchers asked the question '*can optical vortex beams exhibit optical activity with chiral particles analogous to circularly polarized light?*' However, the initial theory and experiments in the field stated that they did not (for a history, see Ref [2]).

2. Present

Around 2018 a flurry of both theoretical and experimental studies appeared which proved quite the contrary [2]. Important discoveries included the unique role that higher-order multipolar moments play in structured light-matter interactions, as well as the importance of non-paraxial optical fields. These works set the foundations for the field of structured light chirality which is now a flourishing and highly topical area of research. The motivation behind undertaking studies in this field is twofold: firstly, engaging the chirality of optical vortices through ℓ expands the scope of chiral light-matter interactions and spectroscopies. For example, before we could use circular dichroism (the differential absorption of circularly polarized light by chiral particles), however a vortex beam produces *vortex dichroism* and, if circularly polarized, *circular-vortex dichroism*. Secondly, chiral light-matter interactions dependent on optical chirality stemming from circular polarization are proportional to σ which is bounded between ± 1. However, a chiral interaction linearly proportional to ℓ has the potential to be significantly enhanced due to the fact ℓ may theoretically take on any integer value to up to ∞.

A remarkable recent result showed that unpolarized light, if an optical vortex mode, exhibits a non-zero optical chirality density (see Fig 2) [3]. What this means physically is that chiral particles will exhibit optical activity with unpolarized light. This polarization independent optical chirality of non-paraxial fields is unique to optical vortices, and therefore structured light.

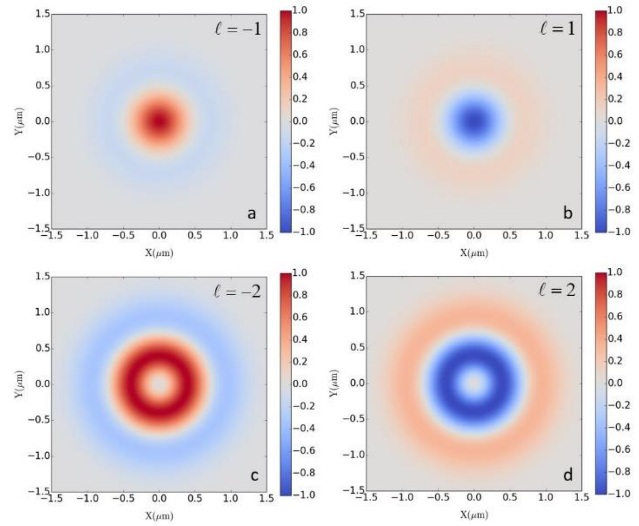

Figure 2: normalized optical chirality density at $w_0 \left(z = 0 \right)$ of an unpolarized Laguerre-Gaussian mode. $p = 0$ in (a)-(d).

3. Future

The cutting-edge includes X-ray [4] and nonlinear methods [5,6], and the field is even expanding beyond optics [7]. Furthermore, there is no doubt that the field will grow into the time-domain through ultrafast chiral methods. The future of structured light chirality looks extremely bright and will continue to push the boundaries of what was thought possible in chiroptical spectroscopy and chiral light-matter interactions.

References

[1] A. Forbes, M. de Oliveira, and M. R. Dennis, *Structured Light*, Nature Photonics **15**, 253 (2021).

[2] K. A. Forbes and D. L. Andrews, *Orbital Angular Momentum of Twisted Light: Chirality and Optical Activity*, J. Phys. Photonics **3**, 022007 (2021).

[3] K. A. Forbes, *Optical Helicity of Unpolarized Light*, Physical Review A **105**, 023524 (2022).

[4] J. R. Rouxel, B. Rösner, D. Karpov, C. Bacellar, G. F. Mancini, F. Zinna, D. Kinschel, O. Cannelli, M. Oppermann, and C. Svetina, *Hard X-Ray Helical Dichroism of Disordered Molecular Media*, Nature Photonics 1 (2022).

[5] K. A. Forbes, *Nonlinear Chiral Molecular Photonics Using Twisted Light: Hyper-Rayleigh and Hyper-Raman Optical Activity*, Journal of Optics **22**, 095401 (2020).

[6] J.-L. Bégin, A. Jain, A. Parks, F. Hufnagel, P. Corkum, E. Karimi, T. Brabec, and R. Bhardwaj, *Scalable Nonlinear Helical Dichroism in Chiral and Achiral Molecules*, (2022).

[7] Q. Tong and S. Wang, *Acoustic Helical Dichroism in a One-Dimensional Lattice of Chiral Resonators*, Physical Review B **105**, 024111 (2022).