Orbital Angular Momentum Transfer in Ro-Vibrational Spectroscopy of Molecules

Georgios M. Koutentakis [1, †], **Mikhail Maslov** [1], **Monika Bahl** [2], **Mateja Hrast** [1], **Tom Jungnickel** [2], **Timo Gaßen** [2], **Oliver H. Heckl** [2], **Mikhail Lemeshko** [2]

[1] Institute of Science and Technology Austria (ISTA), Am Campus 1, 3400 Klosterneuburg, Austria
[2] Christian Doppler Laboratory for Mid-IR Spectroscopy and Semiconductor Optics, Faculty Center for Nano Structure Research, Faculty of Physics, University of Vienna, Boltzmanngasse 5, 1090 Vienna, Austria
†corresponding author's email: georgios.koutentakis@ist.ac.at

Optical vortex beams have intrigued researchers since Allen's seminal 1992 paper [1]. Each photon in these beams carries orbital angular momentum in addition to spin momentum. These beams have diverse applications, including optical tweezers and quantum information processing. Recently, we have shown that the non-zero vorticity of such helical beams can significantly enhance certain ro-vibrational transitions that are otherwise forbidden in non-helical light spectroscopy [2]. In particular, we have found that electric field interacts with the multipole moments of a molecule through its spherical gradients, thus enabling orbital momentum transfer via quadrupole or higher order transitions.

Building on this concept, we aim to optimize the electric field distribution to maximize the interaction between photons and molecules. By analyzing the incident and scattered light by quadrupolar diatomic molecular media, we unveil the field configurations that increase the orbital angular momentum transfer among the light and molecule. Optical vortex lattice configurations, either resulting from either multiple scalar beam interference or vortex beam diffraction, are found to be an ideal platform to observe and control the orbital angular momentum transfer. This method highlights the synergistic interplay between the number, size, and charge of vortex cores, leading to improved interaction efficiency for spectroscopic applications. Harnessing the potential of such structured optical fields, paves the way for significant advances in molecular spectroscopy, potentially providing the means for an extension to mesoscopic objects such as biomolecules or nanoparticles.

Acknowledgments

This research was funded in whole or in part by the Austrian Science Fund (FWF) [10.55776/F1004].

References

[1] L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woerdman, *Phys. Rev. A* **45**, 8185 (1992).

[2] M. Maslov, G. M. Koutentakis, M. Hrast, O. H. Heckl, and M. Lemeshko, *Phys. Rev. Research* **6**, 033277 (2024).