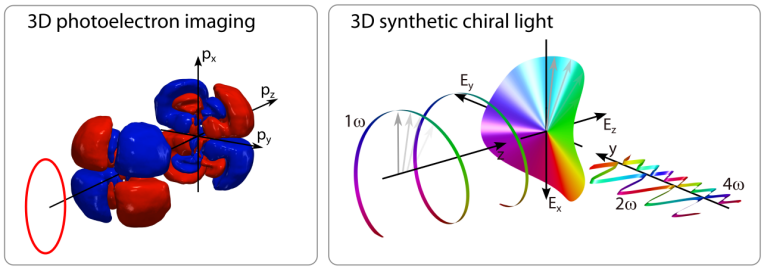
Novel chiral light-matter interaction schemes in the gas phase

Yann Mairesse, CNRS – CELIA, Université de Bordeaux, France
harmodyn.celia.u-bordeaux.fr

Circularly polarized light has been used since the 19th century to distinguish the two mirror images of a chiral molecule, through various forms of circular dichroisms. These processes generally result from non-dipole effects and thus provide weak signals, typically in the 10^{-4}-10^{-3} range. To circumvent this issue, new configurations of chiral light-matter interaction, providing enantio-sensitivity within the electric dipole approximation, have emerged in the past 25 years. They either rely either on increasing the dimensionality of the measurement, for instance by resolving the photoelectron ejection direction in photoelectron circular dichroism [1], or on increasing the dimensionality of the electric field using synthetic chiral light [2].

I will present the results of experiments based on both approaches, using multiple phase-locked femtosecond pulses to control chiral photoionization and laser-induced electron diffraction [3] at the sub-optical-cycle timescale. We will see that resolving the 3D photoelectron momentum distribution enables resolving the handedness of a molecule without imposing the handedness of the light ionizing it, which constitutes a form of non-dichroic enantio-sensitive spectroscopy [4]. On the other hand, we will show that producing a synthetic chiral light field oscillating in 3D enables maximizing the ion yield from a molecule while minimizing that of its mirror image. The branching ratios of different ionic fragments can be controlled in an enantio-sensitive manner by shaping the 3D field, which demonstrates the potential of synthetic chiral light for direct asymmetric photochemistry [5].



[1] N. Böwering, T. Lischke, B. Schmidtke, N. Müller, T. Khalil, and U. Heinzmann, *Asymmetry in Photoelectron Emission from Chiral Molecules Induced by Circularly Polarized Light*, Phys. Rev. Lett. 86, 1187 (2001).
[2] D. Ayuso, O. Neufeld, A. Ordonez, P. Decleva, O. Cohen, M. Ivanov and O. Smirnova, *Synthetic chiral light for efficient control of chiral light–matter interaction*, Nat. Phot. 13, 866 (2019)
[3] D. Rajak, S. Beauvarlet, O. Kneller, A. Comby, R. Cireasa, D. Descamps, B. Fabre, J. D. Gorfinkiel, J. Higuet, S. Petit, S. Rozen, H. Ruf, N. Thiré, V. Blanchet, N. Dudovich, B. Pons, and Y. Mairesse, *Laser-Induced Electron Diffraction in Chiral Molecules,* Phys. Rev. X 14, 011015 (2024)
[4] L. Fede, D. Rajak, C. Sparling, D. Ayuso, V. Blanchet, P. Decleva, D. Descamps, S. Petit, B. Pons, Y. Mairesse, and A. Ordóñez, *Non-Dichroic Enantio-Sensitive Chiroptical Spectroscopy*, arxiv:2512.19062
[5] J.B. Greenwood, L. Fede, D. García González, U. Dellasette, R. Anderson, C. Sparling, D. Townsend, P. Vindel-Zandbergen, D. Ayuso, V. Blanchet, B. Pons, A. Ordóñez, Y. Mairesse, *Direct Asymmetric Photochemistry with Synthetic Chiral Light*, arxiv:2609.12947