

Probing electronic coherences in chiral molecules

Content

At the molecular level, chiral recognition manifests itself through the ability of a chiral entity to distinguish between the two non-superimposable mirror images of another chiral unit, resulting in different physical, chemical and biological properties. The manipulation of these properties requires a deeper understanding of the primary steps of chiral recognition, which is of interest for a wide variety of scientific fields. While signatures of transient molecular chirality have been observed with different chiroptical methods, the manifestation of chiral interactions at the ultrafast electron timescale has not been demonstrated.

Here we use a novel laser technology delivering few-femtosecond (fs) UV pulses [1] to photoexcite chiral methyl-lactate. The molecular relaxation is probed via time-resolved photoelectron circular dichroism (TR-PECD) [2,3] using a time-delayed circularly polarized near-infrared pulse, revealing an ultrafast chiroptical switching effect taking place on a sub-10 fs timescale. By modelling the experiment including both the UV photoexcitation and the photoionization by the circularly polarized probe, we show that the effect is driven by electronic coherences between Rydberg states.

With this experiment we achieve an important milestone of molecular spectroscopy that is to observe and manipulate electronic coherences in neutral molecules. The results demonstrate that TR-PECD provides a route to investigate transient chirality at the electronic timescale with high sensitivity, with numerous applications for a variety of chiral systems such as organometallic complexes and bio-relevant molecules.

[1] Galli M. et al. Optics Letters 44, 1308 (2019)

[2] Comby A. et al. The journal of physical chemistry letters 7, 4514 (2016)

[3] Blanchet V. et al. Physical Chemistry Chemical Physics 23, 25612 (2021)

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