NETLINCS - New Trends in Linear and Non-Linear Spectroscopic Studies of Natural Chirality



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Advances in time-resolved photoelectron circular dichroism of chiral molecules in the gas phase

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PhotoElectron Circular Dichroism (PECD) is an effect driven by electric dipole interactions between an ensemble of chiral molecules and ionizing circularly polarized light [1,2]. It results in a preferential emission direction of photoelectrons along the light propagation axis, leading to an asymmetry that can be observed by recording their angular distribution with an electron detector. Established as a benchmark method to study the static properties of chiral molecules in the gas phase with high sensitivity [3,4], it was extended to the time domain for the investigation of chiral dynamics less than a decade ago [5].

After reviewing the achievements of time-resolved PECD in molecular spectroscopy, I will present the capabilities of our few-femtosecond UV light sources [6,7] to reveal the contribution of ultrafast electron dynamics to the chiral properties of photoexcited neutral molecules [8]. Our results show an ultrafast inversion of the molecular chiroptical response in less than 10 fs after UV excitation, which is captured by modulations of photoelectron angular distributions produced by a time-delayed, circularly polarized ionizing pulse with few-optical-cycle duration. Our theoretical modelling shows that the electronic coherences induced by the UV radiation also generate chiral currents along the molecular structure, offering important prospects for the enantiosensitive, charge-driven reactivity of chiral molecules [9]. As a proof of principle, I will present a case where chiral electron currents can be used as a tool to select the photodissociation direction of chiral molecules with few-fs resolution. Finally, some concluding remarks will be made on the new opportunities made possible by few-femtosecond PECD.

References

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