MO 2: Chirality I

Time: Monday 11:30–13:00 Location: P 110

Invited Talk MO 2.1 Mon 11:30 P 110

Charge-directed reactivity in chiral molecules induced by ultrafast co-rotating currents — •Aurelien Sanchez[1,2,3], Erik Månsson[1], Vincent Wanie[1], Olga Smirnova[4,5], Valerie Blanchet[6], Yann Mairesse[6], Bernard Pons[6], and Francesca Calegari[1,2,3] — [1]Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron DESY, Germany — [2]The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, Germany — [3]Physics Department, Universität Hamburg, Germany — [4]Max-Born-Institut, Berlin, Germany — [5]Technische Universität Berlin, Germany — [6]Université de Bordeaux - CNRS - CEA, CELIA, France

Geometrical phase effects [1] (or the Berry phase [2]) play crucial roles in chemical reactions [3]. Recently, direct evidence of topological effects has been observed in small, isolated chiral structures [4, 5], supported by theoretical work within the electric dipole approximation [6]. One such technique relies on the ability to control chiroptical properties using broadband, short UV pulses to enhance or suppress the anisotropy parameter of a given molecular enantiomer. Here, we extend this method to measure the forward/backward asymmetry of fragments produced by dissociation following ionisation of the neutral excited parent molecule. In doing so, we obtain clean snapshots of the different ion fragments from a particular molecular enantiomer using velocity map imaging (VMI) in combination with short UV pump pulses (FTL 2.1 fs) and near-infrared circularly polarised light (FTL 4.0 fs). At specific delays, we observe a 20 to 30% asymmetry, thereby demonstrating a method for charge-directed reactivity in chiral molecules.

Invited Talk MO 2.2 Mon 12:00 P 110

Controlling the orientational degeneracy of rotational states in chiral molecules — •Elahe Abdiha, Shilpa Yadav, Sejun An, JuHyeon Lee, Boris G. Sartakov, Gerard Meijer, and Sandra Eibenberger-Arias — Fritz-Haber-Institut der Max-Planck-Gesellschaft; Berlin, 14195, Germany

We present recent experimental advances towards full enantiomer-specific control of the quantum states of chiral molecules using enantiomer-specific state transfer (ESST). Early studies on ESST reported only modest state-specific enantiomeric enrichment, limited to a few percent [1,2]. The main limitations are due to the thermal population of rotational states and their spatial degeneracy. Our group has overcome the limitation due to thermal population and achieved near-complete control of the enantiomers in a triad of rotational states containing the absolute ground state [3]. For this, we developed a new approach combining ultraviolet and microwave radiation to remove population from two out of three rotational states before applying ESST. In our recent work, we address the limitation due to spatial degeneracy when applying ESST to rotational states not including the absolute ground state. Here, M-state dependent Rabi frequencies impede the overall transfer efficiency when employing conventional pulse schemes. We incorporate theoretically tailored pulse schemes [4] and demonstrate enhanced control despite degeneracy. [1] Eibenberger et al. Phys. Rev. Lett. 118, 123002 (2017) [2] Pérez et al. Angew. Chem. Int. Ed. 56, 12512 (2017) [3] Lee et al. Nat. Commun. 15, 7441 (2024) [4] Leibscher et al. Commun. Phys. 5, 110 (2022).

MO 2.3 Mon 12:30 P 110

Bottom-up Analysis of Ro-Vibrational Helical Dichroism — •Mateja Hrast, Georgios M. Koutentakis, Mikhail Maslov, and Mikhail Lemeshko — Institute of Science and Technology Austria, Klosterneuburg, Austria

Helical dichroism (HD) is a proposed method for the resolution of molecular chirality, employing the orbital angular momentum (OAM) of light and hypothesized to arise from electric-dipole-electric-quadrupole interactions. Going beyond the conventional assumptions, we propose a general theoretical framework for the analysis of HD, based on molecular symmetries and rotational eigenstates. Using our recently developed model of molecule-light interaction Hamiltonian we establishing an explicit link between chiral resolution and orbital angular momentum (OAM) exchange in light-matter interaction. Tracing microscopic mechanisms of the OAM transfer, we derive the rotational selection rules, which clearly establish that HD emerges only from the spin-orbit coupling of light, even for beams without the far-field OAM. Our findings refine the conditions for observing HD, provide a tool to re-examine the outcome of prior experiments, and guide future designs for chiral sensing with structured light.

MO 2.4 Mon 12:45 P 110

Geometric mechanisms enabling photoelectron spin and molecular chirality coupling in photoionization — •Philip Caesar Flores[1], Stefanos Carlstroem[1], Serguei Patchkovskii[1], Misha Ivanov[1,2,3], Vladimiro Mujica[4], Andres Ordonez[1,5,6], and Olga Smirnova[1,3,7] — [1]Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany — [2]Insitute of Physics, Humboldt University zu Berlin, Berlin 12489, Germany — [3]Technion - Israel Institute of Technology, Haifa, Israel — [4]School of Molecular Sciences, Arizona State University, Tempe, AZ 85287-1604, USA — [5]Department of Physics, Imperial College London, SW7 2BW London, United Kingdom — [6]Department of Physics, Freie Universität Berlin, 14195 Berlin, Germany — [7]echnische Universität Berlin, Straße des 17. Juni 135, 10623 Berlin, Germany

We examine the simplest yet ubiquitous example of spin-chirality coupling: spin-resolved photoionization of randomly oriented chiral molecules, and uncover two universal geometric mechanisms of spin-selective photodynamics, arising solely from electric-dipole interactions and previously unrecognized. These mechanisms are quantified by two pseudovectors stemming from the geometric properties of photoionization dipoles in spin space and in real space. The developed theoretical framework enables rigorous quantification of ‘locking’ between the cation orientation and photoelectron spin, as well the the photoelectron current and its spin. The former effect captures the core of chirality-induced spin selectivity while the latter captures the dynamical origin of spin- and enantio-sensitive multipolar photoelectron currents.