MO 26: Contributions to SYLC II

Time: Thursday 14:30–16:15 Location: P 110

Invited Talk MO 26.1 Thu 14:30 P 110

Differentiating Between Enantiomers with Nuclear Quadrupole Coupling Using Microwave Three-Wave Mixing — •Freya E. L. Berggötz[1,2], Monika Leibscher[3], Wenhao Sun[1], Christiane P. Koch[3], and Melanie Schnell[1,4] — [1]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [2]Institut für Experimentalphysik, Universität Hamburg, Hamburg, Germany — [3]FU Berlin, Fachbereich Physik and Dahlem Center for Complex Quantum Systems, Berlin, Germany — [4]Institut für Physikalische Chemie, CAU zu Kiel, Kiel, Germany

We demonstrate the application of microwave three-wave mixing to the amino alcohol valinol, which displays a hyperfine structure in the rotational spectrum due to nuclear quadrupole coupling. The hyperfine structure complicates the typical triad of rotational states, leading to overlapping microwave three-wave mixing cycles. We identified a set of cycles accessible within the hyperfine substructure of the rotational states $|J_{K_aK_c}\rangle = |1_{01}\rangle, |2_{12}\rangle$, and $|2_{02}\rangle$ by applying the selection rules for rotational transitions and nuclear quadrupole coupling. To address an individual cycle of hyperfine transitions or subsets of cycles simultaneously, we explored different pulse schemes exploiting single-frequency or chirped microwave pulses. Each pulse scheme generated a distinct chiral signal, which shows clear enantiomer differentiation. The experimental findings agree very well with numerical simulations using an effective model for the hyperfine interaction. This study thus extends the applicability of microwave three-wave mixing to previously unexplored molecular systems containing quadrupolar nuclei.

MO 26.2 Thu 15:00 P 110

Dynamical chirality in rotating molecules induced by electron transitions — •Marjan Mirahmadi[1], Alexander Gabriel Löhr[1], Andres Felipe Ordonez[2,3], and Olga Smirnova[1,4] — [1]Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany — [2]Department of Physics, Imperial College London, SW7 2BW London, United Kingdom — [3]Department of Physics, Freie Universit\"at Berlin, 14195 Berlin, Germany — [4]Technische Universität Berlin, Str. des 17. Juni 135, 10623 Berlin, Germany

Although chirality is typically considered a spatial property of an object and depends on its geometrical symmetries such as point inversion and reflection, local chirality arises from a vector tracing a chiral trajectory in time. For instance, locally chiral Lissajous figures of light are known to generate strong enantiosensitive signals in HHG. Inspired by this concept, we introduce locally chiral molecular rotations via rotational wavepackets constructed of few lower rotational states, in contrast to previously demonstrated rotational chirality by using an optical centrifuge.

We then explore the interplay of two locally chiral structures: the induced polarization vector of chiral electron currents and the rotational trajectory of a molecule, both driven by locally chiral fields. We formally investigate how controlled molecular rotation affects the electronic excitations in different ultrafast electronic process. Moreover, including the electron-rotational couplings based on Hund's cases (a) and (b), we provide deeper insight into how electronic currents effect this newly introduced form of rotational chirality.

MO 26.3 Thu 15:15 P 110

Induced Photoelectron Circular Dichroism onto an Achiral Chromophore — •Etienne Rouquet[1,2], Madhusree Roy Chowdhury[1], Gustavo Garcia[1], Laurent Nahon[1], Jennifer Dupont[2], Valéria Lepère[2], Katia Le Barbu-Debus[2], Rodolphe Vuilleumier[3], and Anne Zehnacker[2] — [1]Synchrotron SOLEIL, L'Orme de Merisiers, Départementale 128, F-91190 St Aubin, France — [2]Institut des Sciences Moléculaires d*Orsay (ISMO), CNRS, Université Paris-Saclay, F-91405 Orsay, France — [3]Chimie Physique et Chimie du vivant (CPCV), École Normale Supérieure, Sorbonne Université, CNRS 75005 Paris, France

For the first time, we recorded induced photoelectron circular dichroism (PECD). More specifically, we formed molecular complexes in the gas phase, separated in two moieties. One part is chiral, but does not have a chromophore, and the other is a non-chiral moiety with a chromophore. We recorded induced PECD of the phenol-methyloxirane (Phe-MOx) complex using VUV synchrotron radiation. The HOMO is localized solely on the phenol moiety and we saw a non-zero chiroptical response originating from the ionization of the complex, which we first assigned to the electron scattering off the chiral molecular potential. We then successfully applied this technique using a ns-laser 2 photon energy scheme, providing conformer-selective PECD data. We confirmed our synchrotron-based results on conformer-selected Phe-MOx molecular complexes, with two structures observed in the gas phase. Hydrogen bonding with both lone pairs of the MOx oxygen atom were involved, revealing a chiral deformation of the phenol.

MO 26.4 Thu 15:30 P 110

Enantioselctive Occupation Transfer via Berry Phase Cycling — •Aycke Roos[1], Pablo Maier[1], and Olga Smirnova[1,2] — [1]Max-Born-Institut — [2]Technische Universität Berlin

A chiral molecule interacting with synthetic chiral light acquires geometric properties as the Lissajous figure of this light is adiabatically deformed in a cycle. During this cycle the electronic wave function of the molecule acquires a geometric (Berry) phase.

The phase serves as the fundamental quantity that encapsulates the geometric interplay between field and molecule. It naturally provides a clear geometric picture stripped from any dynamical processes. It further enables the identification of explicit polarization protocols to maximize the enantioselective excitation of one of one enantiomers. In particular, we find that a reversal of the temporal polarization vector trajectory quantified by the winding number leads to a switch between preferential excitation of left-or right handed molecules.

MO 26.5 Thu 15:45 P 110

Enantiosensitive Chern Numbers in Chiral Molecules Driven by Synthetic Chiral Light — •Pablo Maier[1], Aycke Roos[1], and Olga Smirnova[1,2] — [1]Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Straße 2A, 12489 Berlin, Germany — [2]Technische Universität Berlin, Straße des 17. Juni 135, 10623 Berlin, Germany

Temporal geometry describes ultrafast chiral dynamics in molecules by showing how light-driven electronic currents depend on molecular orientation. In our driving scheme the electronic response is effectively independent of one Euler angle, so the relevant orientation space reduces to a sphere parametrized by two angles. The driven electronic states then form smooth bands over this sphere, characterized by a Berry connection, Berry curvature, and associated topological invariants. We show that these orientation-space bands can carry integer-quantized, enantio-sensitive Chern numbers. In a driven three-level electronic manifold used as a minimal model, the dressed eigenstates define three non-degenerate bands whose Chern numbers always sum to zero but are individually nonzero and reverse sign upon exchanging enantiomers. By varying detunings and field parameters, we locate band-touching points where gaps close and the band Chern numbers jump by integer amounts, signaling topological phase transitions between distinct chiral phases.

MO 26.6 Thu 16:00 P 110

Optimising enantioselective electronic population transfer in chiral molecules — •Daniel García González and Andrés Ordóñez — Department of Physics, Freie Universität Berlin, Berlin, Germany

Coherent control offers powerful routes to all-optical enantioselective photochemistry. Our approach is based on electronic transitions, requires neither cold molecules nor intermediate resonances nor chiral sensitisers, and is applicable to randomly oriented molecules. Strong ($> 10\%$) population differences in opposite enantiomers arise from the interference between two multiphoton excitation pathways driven by a fundamental IR field and two of its harmonics.

In this regime, the population difference takes the form [arXiv:2309.02392v2]

$$P_R - P_S \propto \mathrm{Re}\left[\mathbf{g}^{(5)} \cdot \mathbf{h}^{(5)}\right],$$

where $\mathbf{g}^{(5)}$ encodes the molecular geometry, while $\mathbf{h}^{(5)}$ depends on the polarizations of the applied fields and is experimentally controllable.

We study the dependence of $\mathbf{g}^{(5)}$ on the molecular structure using the three conformers of carvone, and introduce a strategy to design polarisation schemes that maximise $\mathrm{Re}[\mathbf{g}^{(5)} \cdot \mathbf{h}^{(5)}]$. This approach yields experimentally realistic field configurations achieving enantioselective population differences of up to 55% -nearly twice the previously reported 30%- offering a clear, geometry-based method for optimising coherent enantioselective control in electronic transitions.