MO 9: Chirality II

Time: Tuesday 11:00–13:00 Location: P 110

Invited Talk MO 9.1 Tue 11:00 P 110

All-Optical Enantiomeric Conversion Observed in a Femtosecond Molecular Movie — •Nicolas Ladda, Fabian Westmeier, Tonio Rosen, Sudheendran Vasudevan, Simon Ranecky, Sagnik Das, Krishna Singh, Hendrike Braun, Jochen Mikosch, Thomas Baumert, and Arne Senftleben — Institute of Physics and CINSaT, University of Kassel, 34132 Kassel, Germany

We present the first observation of the enantiomeric conversion of an optically excited chiral molecule on the femtosecond timescale. Pumping the chiral molecule (methyl p-tolyl sulfoxide) into an electronically excited state with a deep-UV femtosecond laser pulse creates a vibrational wave packet, that can propagate to the other enantiomeric configuration due to a reduced inversion barrier [1]. By measuring time-resolved photoelectron circular dichroism (PECD), these molecular dynamics can be investigated in the gas phase, which provides an interaction-free environment. Advantageously, PECD, i.e., the forward/backward asymmetry of the photoelectron angular distribution with respect to the direction of propagation of ionizing circularly polarized light, is a strong (~10%) effect and can be studied in an energy-resolved manner using a velocity map imaging spectrometer [2].

[1] W. Sun, I. Kleiner, A. Senftleben, M. Schnell, J. Chem. Phys. 2022, 156, 15, 154304.

[2] N. Böwering, T. Lischke, B. Schmidtke, N. Müller, T. Khalil, U. Heinzmann, Phys. Rev. Lett. 2001, 86, 1187

MO 9.2 Tue 11:30 P 110

Photoelectron Circular Dichroism in the Photodetachment of Deprotonated 1-Phenylethanol — •Viktoria Katharina Brandt, André Fielicke, Gerard Meijer, and Mallory Lee Green — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Photoelectron Circular Dichroism (PECD) is a chiroptical effect revealed in the angle-dependent photoemission of an electron from a chiral molecule, upon irradiation by circularly polarized light. PECD can provide insight into our understanding of electron dynamics, as this effect is acutely sensitive to the state of the molecule and the emission conditions of the electron. PECD within the photodetachment of anions has historically been understudied. Past studies of PECD in the photodetachment of anions have reported PECD effects averaged across multiple overlapping electron detachment channels represented by energetically similar tautomers, conformers, and electronic states. In such cases, the sensitivity of PECD to electron dynamics is a double-edged sword: PECD studies can only offer a wealth of information if one can disentangle the individual contributions to the overall measured PECD. Within this study, we measure the PECD effect in the photodetachment of a single electronic state of a single conformer of deprotonated 1-phenylethanol anion to explore the explicit effects of electron kinetic energy and vibrational state on the chiroptical effect within this photoemission regime.

MO 9.3 Tue 11:45 P 110

Robust Prediction of Plasmonic Enhanced Circular Dichroism — •Maria J. Weseloh[1], Mikael Kuisma[2], Tuomas Rossi[3], Xi Chen[4], and Patrick Rinke[1,5] — [1]Aalto University, Finland — [2]Technical University of Denmark, Denmark — [3]CSC - IT Center for Science, Finland — [4]Lanzhou University, China — [5]Technical University of Munich, Germany

Chirality can determine properties such as toxicity, making detection critical for health and environmental safety. Circular dichroism (CD) spectroscopy is a key technique for chiral discrimination but is often limited by weak chiral signals below measurable thresholds. Plasmonic enhancement of molecular circular dichroism by nanoparticles (peCD) enables detection of molecular chirality where conventional CD fails. Real-time time-dependent density functional theory (RT-TDDFT) in principle enables predictive peCD spectra, but in practice a fundamental issue arises: in the traditional length gauge, CD spectra depend on the arbitrary choice of coordinate-system origin - as highlighted in Phys. Rev. B 110, 115423 (2024). We overcome this limitation by implementing a velocity-gauge formulation of RT-TDDFT. Unlike the length gauge approach, where the dipole operator introduces explicit spatial dependence, the velocity gauge employs the momentum operator and removes this spatial dependence. Calculations across 27 distinct origins for molecules and nanoparticle-molecule systems demonstrate a robust origin insensitivity, even with moderately sized basis sets. Thanks to its computational efficiency, our method supports predictive and practical simulations of realistically large systems.

MO 9.4 Tue 12:00 P 110

Photoelectron circular dichroism modified by the Dynamic Stark effect — •Sudheendran Vasudevan[1], Anton N. Artemyev[1], Ignacio M. Casasús[2], Nicolas Ladda[1], Simon T. Ranecky[1], Sagnik Das[1], Tonio Rosen[1], Fabian Westmeier[1], Krishna Kant Singh[1], Daniel M. Haubenreisser[1], Eric Kutscher[1], Jochen Mikosch[1], Hendrike Braun[1], Luis Bañares[2], Philipp V. Demekhin[1], Arne Senftleben[1], and Thomas Baumert[1] — [1]Institute of Physics and CINSaT, University of Kassel, Heinrich-Plett-Strasse 40, 34132 Kassel, Germany — [2]Dpto. de Química Física, Universidad Complutense de Madrid, Avenida de las Ciencias, 28040 Madrid, Spain

Photoelectron circular dichroism (PECD) is a highly sensitive, enantioselective probe of chirality that arises from electric-dipole interactions and appears as a forward/backward asymmetry in photoelectron angular distribution along the light propagation direction. In this work, we investigate how laser intensity affects the PECD of the chiral prototype molecule fenchone using bandwidth-limited near-UV pulses tuned between the 3s and 3p Rydberg states. Increasing intensity alters the Freeman resonances during the 2+1 REMPI process through dynamic Stark shifts and simultaneously reduces the observed PECD. Our time-dependent, single-center model suggests that the dynamic Stark effect mixes an increasingly large set of dipole-coupled virtual states into the effective polarizability of the intermediate state. This admixture of states with differing PECD signatures results in a partial cancellation and a net PECD approaching zero.

MO 9.5 Tue 12:15 P 110

In Situ Circular Dichroism Spectroscopy and Spatial Separation of Chiral Molecules — •Sejun An, Gerard Meijer, and Sandra Eibenberger-Arias — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Chirality is a fundamental symmetry property, yet enantiomers remain difficult to distinguish or control. Enantiomer-Specific State Transfer (ESST) provides a powerful route for enantioselective population control by driving a sequence of microwave transitions [1]. Recent advances in 2024 increased the ESST efficiency to 92%, enabling near-complete state-specific enantiomeric enrichment [2].

Here, ESST is extended toward in situ circular dichroism (CD) spectroscopy. Since the population of a quantum state can be enantioselectively controlled, CD measurements can be performed directly on a racemic mixture without switching laser polarization. This enables CD spectroscopy for chiral molecules that cannot be separated into pure enantiomers by conventional methods, and reduces systematic errors associated with polarization switching, thereby improving reproducibility and sensitivity.

Furthermore, ESST will be further advanced by rotational-state-selective ionization, enabling spatial separation of enantiomers. In this approach, the enantioselectively prepared rotational state is ionized, and the resulting ions can be separated by an external electric field.

[1] S. Eibenberger et al. Phys. Rev. Lett. 118, 123002 (2017)

[2] J. H. Lee et al. Nat. Commun. 15, 7441 (2024)

MO 9.6 Tue 12:30 P 110

Photoelectron Circular Dichroism of Aqueous-Phase Alanine — •Dominik Stemer and Bernd Winter — Fritz Haber Institute of the Max Plank Society, Berlin, Germany

Photoelectron circular dichroism (PECD) manifests as a forward-backward asymmetry in photoelectron flux upon photoionization of chiral molecules by circularly polarized light, and is an exquisitely sensitive probe of molecular chirality, capable of determining enantiomeric excess, but also of resolving differences in molecular conformation and charge state. Although the use of PECD to study chiral biomolecules under physiologically relevant conditions promises to shed light on otherwise difficult-to-access interactions, for example the nature of chirality transfer between a chiral solute and its first solvent shell, the application of PECD to liquid samples is practically challenging. We have previously demonstrated the possibility of measuring PECD for liquid microjets of the neat chiral liquid fenchone, and have more recently confirmed the applicability of PECD to aqueous solutions more generally. Here, we will report on our successful LJ-PECD experiments for core-level photoionization of aqueous-phase alanine, as well as on new results based on the chiral surfactant methylbenzylamine.

MO 9.7 Tue 12:45 P 110

Chiral Discrimination on Gate-Based Quantum Computers — •Sabre Kais[2] and Muhammad Arsalan Ali Akbar[1] — [1]Department of Electrical and Computer Engineering, and Department of Chemistry North Carolina State University, Raleigh, NC 27606 — [2]Department of Electrical and Computer Engineering, and Department of Chemistry North Carolina State University, Raleigh, NC 27606

We present a novel approach to chiral discrimination using gate-based quantum processors, addressing a key challenge in adapting conventional control techniques using modern quantum computing. Schemes such as stimulated rapid adiabatic passage (STIRAP) and shortcuts to adiabaticity (STAP) have shown strong potential for enantiomer discrimination; their reliance on analog and continuous-time control makes them incompatible with digital gate-based quantum computing architectures. Here, we adapt these protocols for quantum computers by discretizing their Gaussian-shaped pulses through Trotterization. We simulate the chiral molecule 1,2- propanediol and experimentally validate this gate-based implementation on IBM quantum hardware. Our results demonstrate that this approach is a viable foundation for advancing chiral discrimination protocols, preparing the way for quantum-level manipulation of molecular chirality on accessible quantum architectures