MO 23: Poster: Chirality

Time: Thursday 17:00–19:00 Location: Tent C

MO 23.1 Thu 17:00 Tent C

Towards perfect enantiomer-specific state transfer of chiral molecules — •Elahe Abdiha, Juhyeon Lee, Johannes Bischoff, Daniel Fontoura Barroso, Boris Sartakov, Gerard Meijer, and Sandra Eibenberger-Arias — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Chiral molecules are important in many chemical and biological processes and are also at the heart of some fundamental physics questions. Recently, enantiomer-specific state transfer (ESST) was experimentally demonstrated [1,2]. Here, the application of three mutually orthogonally polarized microwave fields yields enantiomer-specific population control in a chosen quantum state that is part of a triad of rotational states. I will present our recent work on ESST, where we largely overcome the previous limitation due to initial thermal population by combining ESST with optical methods [3]. By depleting the target state using resonant UV light prior to ESST we achieve state-specific enantiomeric enrichment in the order of 50%. Importantly, we quantitatively study ESST, explicitly including the role of spatial degeneracy. I will also discuss our recent study on the influence of microwave pulse conditions on ESST [4]. Extensions to our scheme will allow to create a molecular beam with an enantiomer-pure rotational level, holding great prospects for future spectroscopic and scattering studies.

[1] Eibenberger et al, PRL 118, 123002 (2017) [2] Pérez et al, Angew. Chem. Int. Ed. 56, 12512 (2017) [3] Lee et al, PRL 128, 173001 (2022) [4] Lee et al, arxiv: 2310.11120 (2023)

MO 23.2 Thu 17:00 Tent C

Intensity dependence of PECD using near-ultraviolet femtosecond laser pulses — •Sudheendran Vasudevan, Han-gyeol Lee, Eric Kutscher, Simon T. Ranecky, Nicolas Ladda, Tonio Rosen, Sagnik Das, Till Stehling, Jayanta Ghosh, Anton N. Artemyev, Hendrike Braun, Arne Senftleben, Philipp V. Demekhin, and Thomas Baumert — Institut für Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel

Exploiting an electric dipole effect in ionization, photoelectron circular dichroism (PECD), i.e., an asymmetry in the photoelectron angular distribution along the light propagation, is a highly sensitive enantioselective spectroscopy for studying chiral molecules in the gas phase using either single-photon [1] or multiphoton ionization [2]. In the latter case, resonance-enhanced multiphoton ionization (REMPI) gives access to intermediate electronic states [3]. The PECD sensitivity opens the door to study control of the coupled electron and nuclear motion in enantiomers. A prerequisite is the detailed understanding of PECD in REMPI schemes. In this contribution, we demonstrate the intensity-dependence of PECD on fenchone via 2+1 REMPI using 30 fs near-ultraviolet laser pulses centered at 396 nm. We find that the magnitude of LPECD decreases with the linear increase in the FWHM of the photoelectron spectra.

References

[1] Böwering, N. et al. Phys. Rev. Lett. 86, 1187 (2001).
[2] Lux, C. et al. Angew. Chem. Int. Ed. 51, 5001*5005 (2012).
[3] Lee, H.-g. et al. PCCP 24, 27483*27494 (2022).

MO 23.3 Thu 17:00 Tent C

Photoelectron circular dichroism after O1s ionization in sec-butyl trimethylsilylether — •Christina Zindel, Emilia Heikura, Catmarna Küstner-Wetekam, Lutz Marder, Johannes Viehmann, Denis Kargin, Rudolf Pietschnig, Andreas Hans, and Arno Ehresmann — Institut für Physik and CINSaT, University of Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Following ionization with circularly polarized radiation, randomly oriented enantiomers show a forward-backwards asymmetry in their photoelectron angular distribution (PAD) with respect to the light's propagation direction. This effect is described as photoelectron circular dichroism (PECD) — a powerful tool to examine the molecular chirality of gas-phase samples. In our studies, we use a velocity map imaging spectrometer (VMI) along with synchrotron radiation to investigate the chiral molecule sec-butyl trimethylsilylether (C_4H_9-O-Si3(CH_3)). When varying the distance between the stereocenter and the silicon atom by subsequently inserting CH_2 groups in between, it is of interest whether the PECD magnitude changes after ionization of the O1s orbital, because although the length of the molecule varies in this case, for oxygen, its geometry with respect to the stereocenter remains the same.

MO 23.4 Thu 17:00 Tent C

Coherent Control of Photoelectron Circular Dichroism using Two-color Laser Pulses — •Till Stehling, Hangyeol Lee, Sudheendran Vasudevan, Simon Ranecky, Nicolas Ladda, Tonio Rosen, Sagnik Das, Jayanta Ghosh, Hendrike Braun, Arne Senftleben, and Thomas Baumert — Universität Kassel Institut für Physik Heinrich-Plett-Str. 40 34132 Kassel/Germany

We study two-pathway interference in multiphoton ionization of fenchone. By employing two pulses at 380 nm and 440 nm, two distinct (2+1) resonance-enhanced multiphoton ionization (REMPI) pathways via the 3s and the 3p band of fenchone are simultaneously driven. The ionization out of these states can result in the same final continuum state, such that the which-way-information is unknown, and interference can be observed. Photoelectron energy-spectra (PES) show three distinct peaks attributed to different ionization pathways. One of which exhibits a dependence on the temporal delay between the two pulses. These preliminary results may indicate a successful implementation of two-pathway interference during the ionization of fenchone. Future experiments will be concerned with the influence of such interference effects on the photoelectron-circular-dichroism (PECD) of fenchone as they present a possible method to implement control schemes in chiral photophysics.

MO 23.5 Thu 17:00 Tent C

Experimental setup to study enhancement of circular dichroism in ion yield of 3-methyl cyclopentanone via tailored femtosecond laser pulses — •Sagnik Das, Jayanta Ghosh, Sudheendran Vasudevan, Hangyeol Lee, Nicolas Ladda, Simon Ranecky, Tonio Rosen, Till Stehling, Fabian Westmeier, Arne Senftleben, Thomas Baumert, and Hendrike Braun — Institut für Physik, Universität Kassel

Femtosecond laser pulses were employed to ionize 3-methyl cyclopentanone (3-MCP) in the home-built time-of-flight mass spectrometer. A 'twin peak' measurement technique [1] was utilized, to obtain ion yields from left and right circularly polarised laser pulses in the same laser shot. Such a technique reduces the statistical error of measurement and allows for simultaneous chirp compensation in both left and right-handed light. Circular dichroism in ion yield (CDIY) is the difference in ion yield from the left and right circularly polarised light for a given enantiomer [2,3]. Switching the enantiomer, the sign of CDIY is flipped. At the wavelengths where we performed the experiments, a 1+1+1 resonance-enhanced multiphoton ionization in 3-MCP takes place via the $\pi^* \leftarrow n$ band, which exhibits a strong circular dichroism. It was found that linear chirp enhances the CDIY. We systematically investigated the reason behind the enhancement of CDIY for chirped pulses.

[1] T. Ring et al., Rev. Sci. Instrum., 92, 033001, 2021
[2] U. Boesl and A. Bornschlegl, ChemPhysChem, 7, 2085, 2006
[3] H. G. Breunig et al., ChemPhysChem, 10, 1199, 2009

MO 23.6 Thu 17:00 Tent C

Time-Resolved Circular Dichroism Spectroscopy with Ultrafast Broadband Circularly Polarized Laser Pulses — •Karina Heilmeier[1], Emely Freytag[2], Christina Kaufmann[2], Frank Würthner[2], Christoph Lambert[2], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Chiral exciton states that have a small dipole strength have only weak contributions in linear absorption spectroscopy. However, such states can be well resolved with circular dichroism spectroscopy. Analogously, time-resolved circular dichroism (TRCD) spectroscopy may give information on the ultrafast dynamics of these exciton states which cannot be resolved with non-chiral transient absorption spectroscopy.[1]

Here, we report a TRCD setup based on the pump–probe approach employing ultrafast broadband white-light probe pulses with left- and right-handed circular polarization. The circular polarization is induced by a polarization grating. In TRCD one measures the difference in absorption of left- and right-circularly polarized light of the excited and non-excited states. To investigate the dynamics of chiral excitons, measurements were carried out on a chiral squaraine dimer and a chiral perylene bisimide dimer, both exhibiting excitonic coupling.

[1] L. Ress, P. Malý, J. B. Landgraf, D. Lindorfer, M. Hofer, J. Selby, C. Lambert, T. Renger, T. Brixner, Chem. Sci. 14, 9328 (2023).

MO 23.7 Thu 17:00 Tent C

Electron correlation in circular dichroism and chirality-induced spin selectivity — •Raoul M. M. Ebeling[1], Maurice Béringuier[1], Vladimiro Mujica[2], Daniel M. Reich[1], and Christiane P. Koch[1] — [1]Freie Universität Berlin, Berlin, Germany — [2]Arizona State University, Arizona, United States of America

We study two phenomena related to the interaction of chiral molecules with circularly polarized light, absorption circular dichroism (CD), and chirality-induced spin selectivity (CISS). We investigate both phenomena in chiral hydrogen and chiral helium, two model systems into which we introduce chirality via an artificial chiral potential. The chiral potential is constructed from a superposition of spherical harmonics and it can be interpreted as a way to mimic the chiral environment of a real molecule. Alternatively, our chiral hydrogen and chiral helium models could even be experimentally realized by placing the atoms in a setup involving several electric fields. Quantifying the chirality of the potential with a suitable measure, we find that it can be tuned via the coefficients in the spherical harmonics expansion. We investigate the influence of the strength of the chiral potential, the strength of the spin-orbit coupling, and the strength of the electron-electron interaction on both CD and CISS.

MO 23.8 Thu 17:00 Tent C

Towards Comparing the PECD Effect in Closed-Shell and Open-Shell Chiral Molecules — •Viktoria K. Brandt, André Fielicke, Gerard Meijer, and Mallory Green — Department of Molecular Physics, Fritz Haber Institute of the MPG, Berlin

Photoelectron Circular Dichroism (PECD) is a chiral optical effect that manifests in the angle-dependent photoemission of an electron upon irradiation of a chiral molecule by circularly polarized light. A PECD effect in anions was observed for the first time in 2021, showing that forces other than long-range coulombic interactions can yield a measurable PECD effect. To further explore the effect of short-range forces, as well as the universal electron dynamics that govern PECD, 1-phenylethanol was investigated as a closed-shell deprotonated anion, a dehydrogenated, neutral radical and a closed-shell neutral. A photoelectron spectrum of the anion showed the presence of only two deprotonated tautomers. However, the landscape of radical isomers, formed by reaction with fluorine atoms, is calculated to be complicated and includes a large number of achiral species, making a comparison to the closed-shell species difficult. In the energy region that overlaps with predicted spectra of chiral isomers, photoionization of the radical enantiomers shows a reversal of a small PECD effect. However, statistics of this effect are low and confirmation of this effect is still inconclusive. At this point, only the closed-shell neutral has a confirmed PECD measured at around 5%. Further work will focus on obtaining a PECD of the closed-shell anion and identifying new molecular targets with less complicated isomeric landscapes.

MO 23.9 Thu 17:00 Tent C

Raman Optical Activity Of Glucose — •Klaus Hofmann, Luisa Martin, and Ingo Fischer — Universität Würzburg, 97074 Würzburg, Germany

Raman Optical Activity (ROA) is a type of vibrational circular dichroism: chiral samples show different Raman intensities when utilizing circular polarized light. The ROA signal is very sensitive to the molecular geometry and environment of the sample, but also exhibits high levels of noise and false signals, since the intensity difference is roughly 0.1% of the corresponding Raman peak.

Using a custom built cost-efficient ROA spectrometer, the signal of the two glucose enantiomers in aqueous solution was recorded, with exposure times of one week each. Python was used to automate the setup modulation, data acquisition and post-processing. The result agrees with literature and matches spectra acquired on commercially availible spectrometer.