MO 23: Contributions to SYLC I

Time: Thursday 11:00–12:45 Location: P 110

Invited Talk MO 23.1 Thu 11:00 P 110

Large circular dichroism in the total photoemission yield of free chiral nanoparticles created by a pure electric dipole effect — •Sebastian Hartweg[1,2], Dušan K. Božanić[3], Gustavo A. Garcia-Macias[2], and Laurent Nahon[2] — [1]Institute of Physics, University of Freiburg, Germany — [2]Synchrotron Soleil, St. Aubin, France — [3]Vinča Institute of Nuclear Sciences, University of Belgrade, Belgrade, Serbia

Spectroscopic techniques that are sensitive to molecular chirality are important analytical tools to quantitatively determine enantiomeric excess and purity of chiral molecular samples. Many chiroptical processes however produce weak enantio-specific asymmetries due to their origin relying on weak magnetic dipole or electric quadrupole effects. Photoelectron circular dichroism (PECD) in contrast, is an intense effect, that is fully contained in the electric dipole description of light matter interaction and creates a chiral asymmetry in the photoelectron angular distribution. Here, we demonstrate that this chiral signature in the angular distribution of emitted electrons can be translated into the total photoionization yield for submicron-sized condensed samples. The resulting chiral asymmetry of the photoionization yield (CAPY), mediated by the attenuation of light within the particles, can be detected experimentally without requiring high vacuum systems and electron spectrometers. This effect can be exploited as an analytical tool with high sensitivity to chirality and enantiopurity for studies of chiral organic and hybrid submicron particles in environmental, biomedical or catalytic applications.

MO 23.2 Thu 11:30 P 110

Investigating transient localized charges in small chiral molecules with free-electron lasers — •Markus Ilchen on behalf of a large collaboration — University of Hamburg, Germany — Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — European XFEL, Schenefeld, Germany

Chirality, the inherent "handedness" of many molecular structures, is a cornerstone of biological processes, influencing and steering interactions at the molecular level as well as shaping a broad variety of the physical and chemical properties of matter. The technological evolution of free-electron lasers will be presented in terms of polarization control and high-power attosecond pulse generation in the soft X-ray regime, which enables them to create and explore transiently chiral systems with localized charges at the onset of structural dynamics. This opens the new field of site-specific investigations of building blocks of life at the onset of dynamics outside of the nucleus.

MO 23.3 Thu 11:45 P 110

Correlation between 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 investigate whether absorption circular dichroism (CD) and chirality-induced spin selectivity (CISS) share common physical origins in the interaction of chiral molecules with circularly polarized light. Using a one-electron model in which an effective chiral potential mimics a chiral molecule, we analyze how the chiral potential and spin-orbit coupling strength influence CD and CISS, respectively. We find that the CD signal increases with the chiral potential strength and decreases with stronger spin-orbit coupling. The difference in spin polarization shows a more complex behavior, and we surprisingly find that strong spin-orbit coupling or strong chiral potentials reduce its value. This is caused by the corresponding Hamiltonian terms diminishing the effect of the other, a trend clearly reflected in the energy scales of the competing interactions. Consequently, the strongest enantiomer-sensitivity of the spin polarization is obtained when balancing the two. Lastly, we observe a correlation between CD and CISS that becomes weaker for strong chiral potentials.

MO 23.4 Thu 12:00 P 110

Non-Dichroic Enantio-Sensitive Chiroptical Spectroscopy — Letizia Fede[1], Debobrata Rajak[2], Chris Sparling[3], David Ayuso[4], Valérie Blanchet[1], Piero Decleva[5], Dominique Descamps[1], Stéphane Petit[1], Bernard Pons[1], Yann Mairesse[1], and •Andrés Ordóñez[6] — [1]Université de Bordeaux CNRS CEA CELIA, France — [2]ELI ALPS, Hungary — [3]Heriot-Watt University, UK — [4]Imperial College London, UK — [5]Università degli Studi di Trieste, Italy — [6]Freie Universität Berlin, Germany

Chiroptical effects using circularly polarized light produce signals that change sign when switching either molecular handedness (enantiosensitivity) or light helicity (circular dichroism). Here, we break this enantiosensitive-and-dichroic paradigm by measuring a new type of chiroptical signal which is enantiosensitive but not dichroic. We photoionize chiral molecules using a strong laser field and detect the three-dimensional photoelectron momentum distribution (PMD). The Non-Dichroic, Enantio-Sensitive asymmetry is encoded in octupolar and higher multipolar terms in the PMD, appearing in multiphoton ionization with elliptical or cross-polarized two-color fields. Our simulations agree with our measurements and show that this effect extends from the two-photon to the strong-field ionization regime. The robustness of the enantiosensitivity with respect to the relative phase between the orthogonal components of the ionizing field represents an example of symmetry protection, opening unexplored opportunities for imaging ultrafast dynamics in chiral molecules, such as enantiosensitive photoelectron spectroscopy with bright squeezed vacuum states.

MO 23.5 Thu 12:15 P 110

Femtosecond transient circular dichroism spectroscopy of chiral organic-inorganic lead perovskites — •Thomas Lenzer and Kawon Oum — University of Siegen, Physical Chemistry 2, Adolf-Reichwein-Str. 2, 57076 Siegen

Chiral lead-based perovskites represent promising candidates for the generation of spin-polarized charge carriers, enabling advances in spintronic and optoelectronic applications. We report thin films of hybrid organic-inorganic perovskites that exhibit pronounced circular dichroism (CD) responses reaching 5000 mdeg and dissymmetry factors exceeding 0.10, with negligible contributions from linear dichroism and linear birefringence. Using transient circular dichroism (TrCD) spectroscopy with femtosecond resolution,[1,2] we investigate the ultrafast dynamics of spin-polarized carriers. Complementary transient absorption measurements reveal the dominant relaxation pathways following photoexcitation in these materials.

[1] D. Gust, M. Scholz, V. Schumacher, J.-C. Mulatier, D. Pitrat, L. Guy, K. Oum and T. Lenzer, Sci. Rep. 2024, 14, 12694. [2] M. Morgenroth, M. Scholz, M. J. Cho, D. H. Choi, K. Oum and T. Lenzer, Nat. Commun. 2022, 13, 210.

MO 23.6 Thu 12:30 P 110

Ultra-fast nonlinear optical response of chiral molecules with a focus on conformer sensitivity — •Elena Aethra Christou[1,3], David Ayuso[2], Felipe Morales[1], and Olga Smirnova[1,3] — [1]Max Born Institute, Berlin, 12489, Germany — [2]Imperial College London, SW7 2AZ,UK — [3]Technische Universität Berlin, 10623, Germany

We model the ultrafast nonlinear response of chiral molecules using TDDFT and temporally and spatially confined, CEP-controlled linearly polarized pulses. Focusing generates a longitudinal field component that produces forward elliptically polarized light, which in turn drives a chiral response orthogonal to the propagation plane, leading to even-harmonic emission. Interference between achiral odd and chiral even harmonics, shaped by the CEP, yields enantio-sensitive optical rotation. Building on previous work demonstrating this, we extend this approach to investigate the interplay between electronic dynamics and molecular conformations. We apply this mechanism to the three most abundant conformers of the chiral amino acid serine, which have reported gas-phase populations of 43.7%, 18.8%, and 14.8%. Their distinct polarization signatures are quantified through molecular QR codes that encode conformer-dependent chiral dynamics. These differences reveal a clear conformer-specific chiral response. The approach demonstrates the potential of CEP-controlled fields for conformer-resolved chiral spectroscopy in the near-infrared regime.