SYCU 1: Chirality meets Ultrafast

Time: Monday 11:00-13:00

Location: e415

Invited TalkSYCU 1.1Mon 11:00e415Photoelectron circular dichroism in the light of resonance enhanced multi-photon ionization — •THOMASBAUMERT — Universität Kasselversität Kassel / D-34132Kassel

Exploiting an electric dipole effect in ionization [1], photoelectron circular dichroism (PECD) is a highly sensitive enantiospecific spectroscopy for studying chiral molecules in the gas phase using either single-photon ionization [2] or multi-photon ionization [3]. In the latter case resonance enhanced multi-photon ionization (REMPI) gives access to neutral electronic excited states. The PECD sensitivity opens the door to study control of the coupled electron and nuclear motion in enantiomers. A prerequisite is a detailed understanding of PECD in REMPI schemes. In this contribution I will report on our recent experiments devoted to unravel different aspects of this effect on the fenchone prototype by addressing the range from impulsive excitation on the femtosecond time scale to highly vibrational state selective excitation with the help of high resolution nanosecond laser techniques. The reflection of the number of absorbed photons in the PECD will be discussed as well as subcycle effects in bichromatic fields.

[1] B. Ritchie, Phys. Rev. A 1976, 13, 1411-1415.

[2] N. Böwering, T. Lischke, B. Schmidtke, N. Müller, T. Khalil, U. Heinzmann, Phys. Rev. Lett. 2001, 86, 1187-1190.

[3] C. Lux, M. Wollenhaupt, T. Bolze, Q. Liang, J. Köhler, C. Sarpe, T. Baumert, Angew. Chem. Int. Ed. 2012, 51, 5001-5005.

Invited Talk SYCU 1.2 Mon 11:30 e415 New strategies for controlled chirality from the rovibrational dynamics of molecules — •ANDREY YACHMENEV — Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607, Hamburg, Germany

Chirality is a subject of recurrent interest across many fields of science, from the parity violation effects in fundamental physics to the enantiomeric recognition in biochemical reactions and ultimately the evolution of terrestrial life and the origins of homochirality. The development of experimental methods to study chiral molecular phenomena is therefore very important. In recent years, a number of groundbreaking techniques have been proposed and realised providing far more sensitive probes of molecular chirality than traditional chiroptical methods [1-4], and that at the ultrafast time-scales.

In my talk, I will present alternative approaches to ultrafast probes of molecular chirality, that are based on the strong-field controlled rovibrational dynamics of molecules. In particular, I will focus on the methods for inducing dynamic chirality in statically achiral molecules [5], efficient spatial separation of chiral molecules [6], as well as detecting the enantiomeric excess.

[1] O. Neufeld, et al., Phys. Rev. X 9, 031002 (2019)

[2] D. Baykusheva, et al., PNAS 116, 23923 (2019)

[3] S. Beaulieu, et al., New. J. Phys. 18, 102002 (2016)

[4] M. Pitzer, et al., Science 341, 1096 (2013)

[5] A. Owens, et al., Phys. Rev. Lett. 121, 193201 (2018)

[6] A. Yachmenev, et al., Phys. Rev. Lett. 123, 243202 (2019)

Invited Talk

 $\mathrm{SYCU}~1.3 \quad \mathrm{Mon}~12{:}00 \quad \mathrm{e}415$

Time-dependency in Photoelectron Circular Dichroism: from femtosecond scale to attosecond — •VALERIE BLANCHET — Centre des Lasers Intenses et Applications — University of Bordeaux, Talence, France

Life is a chiral puzzle based on the stereochemistry of chiral molecules and chiral receptors. Tackling this chiral recognition at the atomic scale by taking into account the electronic cloud is one of the Grails of chemical physics. Meanwhile photoionization can produce strong chiral signals : when photoionization of chiral molecules is induced by circularly polarized light, the 3D-angular distribution of photoelectrons exhibits a forward/backward asymmetry with respect to the laser propagation axis. This effect has been called PhotoElectron Circular Dichroism (PECD) and results from the scattering of the electrons in the chiral molecular potential. It is quantum mechanically described as interferences between partial ionization waves. In this talk, time-resolved PECD will be presented. But is there a temporal correspondence between the interference nature of PECD and the photoemission times of these electrons? We answer this question by using two different experimental setups : a RABBIT (Reconstruction of Attosecond Beatings By Interference of Two-photon transitions) ionization technique with chiral light to which we refer to as CHABBIT and a two-color ionization scheme leading to an Enantiosensitive Sub-Cycle Antisymmetric Response Gated by electric field rOTation (ESCARGOT). In ESCAR-GOT, we control the instantaneous ellipticity and chirality of the light at the sub-cycle level.

Chiral discrimination is hard, especially on ultrafast time scales. Standard optical methods rely on the molecules feeling the helix that circularly polarized light creates in space. However, the pitch of this helix is much larger than the molecules, and chiral response is weak.

We have recently introduced synthetic chiral light [1], a new type of freely-propagating optical fields in which the tip of the electric field vector draws a chiral, three-dimensional Lissajous curve in time, at each fixed point in space. Such light is locally chiral: unlike circularly polarized light, its chirality does not rely on the spatial helix of the light field. Thus, it remains chiral in the dipole approximation. Here I will show how to generate such light practically, how to characterize and control its handedness, and how to maintain it globally, across the entire interaction region. Synthetic chiral light enables the highest possible degree of control over the enantio-sensitive nonlinear optical response at the level of total signal intensities, opening efficient ways for imaging and controlling chiral matter on ultrafast time scales.

[1] D. Ayuso et al, Nat. Phot. 13, 866 (2019)