

## Symposium Chirality meets ultrafast (SYCU)

jointly organized by  
the Molecular Physics Division (MO) and  
the Quantum Optics and Photonics Division (Q)

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Chiral molecules, including elementary building blocks of life such as amino acids, are molecules which are not superimposable on their mirror images. Their chiral structures, and the chiral complexes they form, facilitate molecular recognition. It is this chiral recognition which enables our senses of smell and taste. Chiral recognition is also crucial in pharmaceutical applications – it allows us to metabolize drugs. This is why understanding chiral interactions and distinguishing different enantiomers of a chiral molecule are such important problems with applications in physics, chemistry, biology, and medicine.

We are currently witnessing a revolution in our ability to use light to discriminate chiral molecules and to resolve the chiral electronic and nuclear dynamics underlying chiral functions. Since the times of Louis Pasteur in the XIX-th century, molecular chiral recognition with light relied on weak interaction between the molecule and the magnetic-field component of the light wave. This symposium will spotlight a disruptive step, the electric-dipole revolution, in chiral discrimination, which enabled the extremely efficient interaction between light and chiral matter. This opened the way not only to ultrafast imaging, but also to controlling such interactions in chiral molecules on various time scales, from electronic and vibronic to rotational, and eventually even to spatially separate enantiomers. Efficient control over chiral light-matter interactions requires a new type of light, i.e., a synthetic chiral electric field, which is fundamentally different from the circularly polarized light we are familiar with.

## Overview of Invited Talks and Sessions

(Lecture hall Audimax)

### Invited Talks

MO 3.1	Tue	10:45–11:15	H3	<b>Photoelectron circular dichroism in the light of resonance enhanced multi-photon ionization</b> — ●THOMAS BAUMERT
SYCU 1.1	Tue	14:00–14:30	Audimax	<b>Overview of the temporal dependencies of Photoelectron Circular Dichroism</b> — ●VALERIE BLANCHET
SYCU 1.2	Tue	14:30–14:45	Audimax	<b>Ultrafast, all-optical, and highly enantio-sensitive imaging of molecular chirality</b> — ●DAVID AYUSO
SYCU 1.3	Tue	14:45–15:00	Audimax	<b>Hyperfine interactions in rotational chiral states</b> — ●ANDREY YACHMENEV
SYCU 1.4	Tue	15:00–15:30	Audimax	<b>Chiral molecules in an optical centrifuge</b> — ●VALERY MILNER, ALEXANDER MILNER, ILIA TUTUNNIKOV, ILYA AVERBUKH
SYCU 1.5	Tue	15:30–16:00	Audimax	<b>Enantiomer-selective controllability of asymmetric top molecules</b> — ●MONIKA LEIBSCHER

### Sessions

SYCU 1.1–1.5 Tue 14:00–16:00 Audimax **Chirality meets ultrafast**

### Session about chirality within MO

MO 3.1–3.8 Tue 10:45–13:00 H3 **Chirality**

## SYCU 1: Chirality meets ultrafast

Time: Tuesday 14:00–16:00

Location: Audimax

**Invited Talk** SYCU 1.1 Tue 14:00 Audimax  
**Overview of the temporal dependencies of Photoelectron Circular Dichroism** — ●VALERIE BLANCHET — CELIA, university of Bordeaux/CNRS/CEA

In this talk, different experimental approaches of time-resolved photoelectron asymmetry of chiral molecules will be presented ranging from attosecond resolution to a femtosecond one.

**Invited Talk** SYCU 1.2 Tue 14:30 Audimax  
**Ultrafast, all-optical, and highly enantio-sensitive imaging of molecular chirality** — ●DAVID AYUSO — Department of Physics, Imperial College London, London, United Kingdom — Max-Born-Institut, Berlin, Germany

Chiral molecules appear in pairs of opposite mirror twins called enantiomers, which behave identically unless interacting with another chiral object. Distinguishing them is vital, but also hard. Traditional optical methods rely on the molecules feeling the spatial helix of circularly polarized light. However, the pitch of this helix is too large, leading to weak enantio-sensitivity and making chiral discrimination difficult, especially on ultrafast time scales. In other words, the enantio-sensitive component of the optical response is weak because it arises beyond the electric-dipole approximation.

In this talk, I will present new ways of imaging molecular chirality using tailored light. I will show how, by shaping the Lissajous figure that the tip of the electric-field vector of the laser draws in time, we can make the nonlinear response of chiral molecules highly enantio-sensitive, already in the electric-dipole approximation. A key ingredient of our recipe is the longitudinal field component that arises in a non-collinear geometry [Ayuso et al, Nat Photon 13, 866 (2019)] or in a tightly focused beam [Ayuso et al, arXiv:2011.07873 (2020)]. The possibility of generating strongly enantio-sensitive signals via purely electric-dipole interactions creates new opportunities for highly enantio-sensitive imaging and control of molecular chirality and ultrafast chiral dynamics.

**Invited Talk** SYCU 1.3 Tue 14:45 Audimax  
**Hyperfine interactions in rotational chiral states** — ●ANDREY YACHMENEV — Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85 (building 49a, room 203), 22607 Hamburg

Rotational chirality is a phenomenon associated with spontaneous symmetry breaking by unidirectionally spinning molecules into so-called rotational-cluster states [1,2]. These states are characterized by strong centrifugal forces pulling the symmetry-equivalent atoms in the molecule into different directions, thus breaking permutation symmetry. Here, we consider the effects of nuclear-spin hyperfine mixing in rotationally chiral states. For the H<sub>2</sub>S molecule we demonstrated, through comprehensive variational calculations, strong ortho-para state mixing in rotational cluster states leading to the generation of spin polarization and magnetization.

[1] P. R. Bunker and P. Jensen, *J. Mol. Spectrosc.* 228, 640 (2004)  
 [2] A. Owens, A. Yachmenev, S. N. Yurchenko, J. Küpper, *Phys. Rev. Lett.* 121, 193201 (2018)

**Invited Talk** SYCU 1.4 Tue 15:00 Audimax  
**Chiral molecules in an optical centrifuge** — ●VALERY MILNER<sup>1</sup>, ALEXANDER MILNER<sup>1</sup>, ILIA TUTUNNIKOV<sup>2</sup>, and ILYA AVERBUKH<sup>2</sup> — <sup>1</sup>Department of Physics & Astronomy, The University of British Columbia, Vancouver, Canada — <sup>2</sup>Department of Chemical and Biological Physics, The Weizmann Institute of Science, Rehovot, Israel

The fundamental importance of molecular chirality drives the search for novel techniques of enantioselective control, detection, and separation of chiral molecules. It has been recently predicted that an optical centrifuge - a laser pulse, whose linear polarization undergoes an accelerated rotation around its propagation direction, may orient chiral molecules in the laboratory frame, with the orientation axis dependent on the handedness of the enantiomer. Here we present the first experimental observation of this phenomenon [1,2] in propylene oxide molecules. The demonstrated technique offers not only an alternative way of differentiating between molecular enantiomers, but also a new approach to enantioselective manipulation of chiral molecules with light. We discuss the efficiency of the method and the ways to improve it further with new techniques of rotational excitation and detection of the directional molecular rotation.

[1] A. A. Milner, J. Fordyce, I. MacPhail-Bartley, W. Wasserman, V. Milner, I. Tutunnikov and I. Sh. Averbukh, *Phys. Rev. Lett.* 122, 223201 (2019).

[2] I. Tutunnikov, J. Floß, E. Gershnel, P. Brumer, I. Sh. Averbukh, A. A. Milner and V. Milner, *Phys. Rev. A* 101, 021403(R) (2020).

**Invited Talk** SYCU 1.5 Tue 15:30 Audimax  
**Enantiomer-selective controllability of asymmetric top molecules** — ●MONIKA LEIBSCHER — Theoretische Physik, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany

Three-wave mixing (3WM) spectroscopy of chiral molecules allows for enantio-selective population transfer despite random orientation of the molecules [1]. Three-wave mixing is usually regarded as a three-state process [2]. However, due to the M-degeneracy of the rotational spectrum, the underlying rotational dynamics is far more complex and the usual 3WM schemes do not reach complete selectivity.

We prove complete controllability for rotational states of an asymmetric top molecule and introduce the concept of enantiomer-selective controllability. This allows us to determine the conditions for complete enantiomer-specific population transfer in chiral molecules and to construct pulse sequences realizing this transfer for population initially distributed over degenerate M-states [3].

[1] D. Patterson, M. Schnell, and J. M. Doyle, *Nature* 497, 475 (2013). [2] M. Leibscher, T. F. Giesen, C. P. Koch, *J. Chem. Phys.* 151, 014302 (2019). [3] M. Leibscher, E. Pozzoli, C. Perez, M. Schnell, M. Sigalotti, U. Boscain, and C. P. Koch, arXiv:2010.09296 (2020).