

NETLINCS - New Trends in Linear and Non-Linear Spectroscopic Studies of Natural Chirality

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Book of Abstracts

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Theory / 2**A perspective on chiral structured light****Author:** Kayn Forbes¹¹ *University of East Anglia***Corresponding Author:** k.forbes@uea.ac.uk

Since the pioneering work by French scientists in the early 1800s, optical activity and chiral light-matter interactions have been produced via the chirality of light stemming from a degree of ellipticity in its local polarisation state. The mechanism is well understood: the polarisation state can rotate in a left or right-handed fashion and leads to differential interactions with left and right-handed materials. Structured light can be chiral in degrees of freedom beyond local polarisation state. Optical vortices, for example, possess a chiral wavefront (a twisting tornado structure) due to their azimuthal phase. This chirality is completely independent of that associated with the local state of polarisation. Early studies in the 2000s concluded this chirality of optical vortices can play no role in chiral light-matter interactions. Fast forward to 2018 and all of this changed, with multiple studies proving it was in fact possible. This talk aims to lay out a 'past, present, and future' perspective on this rapidly emerging field of 'chiral structured light'. We will provide a narrative of the field of research starting from its origin in those initial negative results all the way through to experiments highlighting the new and enhanced chiral light-matter interactions provided by structured light. Along the way we will develop and present the underlying theoretical breakthroughs. We will present and discuss a selection of current start-of-the-art results in the area, and conclude with some thoughts for future endeavours.

Gas Phase Studies / 3**Recent results on static valence-shell Photo-Electron Circular Dichroism (PECD) on gas phase systems****Author:** Laurent Nahon¹¹ *Synchrotron SOLEIL***Corresponding Author:** laurent.nahon@synchrotron-soleil.fr

Within a bottom/up approach of molecular complexity, the study of isolated, substrate- and solvent-free chiral species is crucial, but classical chiroptical probes such as Circular Dichroism (CD) in absorption are poorly adapted to dilute matter because of their very weak associated asymmetries. At the opposite, two decades ago was introduced a new chiroptical effect called Photoelectron Circular Dichroism (PECD), allowed in the electric dipole approximation, leading to very intense (up to 40 %) forward/backward asymmetries, with respect to the photon axis, in the angular distribution of photoelectrons produced by circularly polarized light ionization of gas phase pure enantiomers. PECD happens to be a universal, orbital-specific, photon energy dependent chiroptical effect and is a subtle probe of the molecular potential, on which the photoelectron is scattered off, being very sensitive to static molecular structures such as conformers, isomers, clusters, as well as to vibrational motion, much more so than other observables in photoionization such as the cross section (Photoelectron Spectrum-PES) or the usual (achiral) χ asymmetry parameter (for a review see [1]). Therefore, PECD studies have a dual fundamental and analytical interest. This last aspect is probably the driving force for the recent extension and blooming of PECD studies, beyond the pioneering synchrotron radiation works, towards the laser (and now FEL) community.

After a broad introduction to PECD, several recent results regarding static one VUV-photon valence-shell PECD will be presented, including a striking example of the specific sensitivity of PECD to conformations [2] as well as a first evidence of induced-PECD onto an achiral chromophore within a molecular complex [3]. Then the demonstration of conformer-specific PECD via two-photon ns-laser ionization will be highlighted [4].

The conclusion will stress the universality of PECD, which may be at play in numerous environments including the interstellar medium where it has been suggested as a possible symmetry-breaking process in link with the origin of life's homochirality [5].

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Welcome / 7

Welcome to the NETLINCS Workshop and Introductory Remarks

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High Resolution Spectroscopy and Molecular Chirality: Symmetries and primary processes between less than yoctoseconds and more than days

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To be announced

Corresponding Author: winter@fhi-berlin.mpg.de

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Electron dynamics in chiral semiconductors

Author: Alberto Crepaldi¹

¹ *Politecnico di Milano*

Corresponding Author: alberto.crepaldi@polimi.it

The growing interest in chirality represents a notable example of *convergence* between different areas of research within the realm of condensed matter. The observation of skyrmions in compounds of group B20 [1] represents only the first report of a wealth of magnetic ordering [2] and collective excitations [3] that can be realized only in the absence of inversion and mirror symmetry operations. Topology predicts the existence of new Weyl fermions [4], and chiral symmetry is responsible for unique spin arrangements in momentum space [5, 6], along with large Berry curvature that is key ingredient for non-linear optical [7] and transport properties [8].

Elemental tellurium is one of the simplest chiral crystals and it is an ideal playground to study the

interplay between symmetries and other interesting physical properties. In my talk I will discuss how intense laser light can be used to excite coherent phonons, both total symmetric Ag mode and symmetry breaking Eg mode. By using time and angle-resolved photoelectron spectroscopy (trARPES) we can track the dynamical change in the band structure, and the comparison with state-of-the-art TDDFT+U calculation reveals the microscopic origin of the in-phase oscillations of the edge of the band-gap, due to phonon-induced modulation of the effective Hubbard U term [9]. All optical techniques, including second (and higher order) harmonic generation provide complementary information about the light-induced change in symmetry, which indicates a promising route to alter the topological phase and the spin texture in Te.

Finally, I will give an overview of our activity on other chiral semiconductors, CdAs₂ and (TaSe₄)₂I [10], in which intense laser light is responsible for a change in the electronic population that persists on time scale ranging from microsecond to minutes, of potential interest for opto-electronics and light harvesting based on the bulk photovoltaic effect [11].

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To be announced

Corresponding Author: ravi.bhardwaj@uottawa.ca

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Core-level photoelectron spectroscopy for the investigation of ultrafast chiral dynamics in molecules

Author: Caterina Vozzi¹

¹ CNR-IFN

Corresponding Author: caterina.vozzi@cnr.it

A molecule is chiral if it lacks both a plane and a center of symmetry, resulting in two non-superimposable mirror-image forms known as enantiomers. Chirality is crucial in the physical and biological sciences because enantiomers, despite having the same chemical structure, often exhibit different chemical and physical properties when interacting with other chiral entities. While chirality is extensively studied in biology, chemistry, and pharmaceuticals, the ultrafast dynamics of chiral compounds remain largely unexplored. A promising technique for investigating these dynamics is Time-Resolved Photo-Electron Circular Dichroism (TR-PECD), which uses an ultrashort circular pulse to ionize the molecule from its electronically excited states, revealing time-dependent dichroism through the asymmetry in photoelectron angular distribution. However, interpreting TR-PECD experiments is challenging due to the non-local nature of the initial excited state.

Recently, Time-Resolved X-ray Photoelectron Spectroscopy (TR-XPS) has shown potential in probing local relaxation dynamics of electronically excited states with chemical specificity. TR-XPS can distinguish different atomic sites by analyzing their transient excited-state chemical shifts.

Recently, we combined the chemical and site-specificity of TR-XPS with the enantio-sensitivity of TR-PECD to study the relaxation dynamics of photoexcited fenchone molecules at the carbon K-edge. This experiment demonstrates that femtosecond chiral dynamics can be effectively probed using core-level spectroscopy with circularly polarized XUV light provided by the FERMI free-electron laser. Additionally, core-level PECD spectroscopy of transient excited states enables us to isolate different carbon atoms within the molecule through their distinct chemical shifts, enhancing the site-specificity of the technique. This allows for separating and identifying PECD from various C-atom sites in the molecule that would otherwise be inaccessible.

Furthermore, we will discuss preliminary results of other experiments performed at FERMI and XFEL to extend this approach to other relevant ultrafast chiral dynamics.

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Mueller matrix polarimetry for chiral detection in the solid state

Author: Oriol Arteaga¹

¹ UB

Corresponding Author: oarteaga@ub.edu

This presentation will review our recent progress studying chiral media with Mueller matrix polarimetry. A comprehensive presentation of our polarimetric method will be provided, detailing the instrumentation used, a few remarks about data analysis as well as some basic concepts about light propagation in chiral anisotropic media. We will discuss how optical characterization methods based only on circularly polarized light can often render inconclusive or erroneous results for chiral assessment. We will show that Mueller matrix spectroscopy and imaging techniques are suitable characterization tools to unveil the ever-growing complexity of anisotropic chiral media. Mueller matrix polarimetry allows the measurement of circular dichroism (CD) together with other optical effects, e.g. linear birefringence, linear dichroism, and circular birefringence. These measurements are critical when studying the optical activity of supramolecular assemblies, nanomaterials, or, more in general, any solid-state system. Examples of chiroptical spectroscopy and chiroptical imaging based on complete polarimetry will be provided.

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Twist and Shine: Helical Zone Plates for Probing Chirality

Author: Benedikt Rösner¹

¹ Paul Scherrer Institut

Corresponding Author: benedikt.rosner@psi.ch

This presentation will give an overview about the generation and applications of X-ray beams that exhibit an **orbital angular momentum** (OAM) using helical zone plates, focusing particularly on their ability to probe chiral properties in matter. Helical (or spiral) zone plates represent an advanced class of diffractive optical lenses that can modulate the phase of light while focusing it at the same time. The resulting beams with twisted wavefronts carry OAM, a property of light that is different from the **spin angular momentum** (SAM) that is widely known from X-ray circular dichroism.

In the first part of the presentation, we will hear about the principle on OAM-carrying radiation and shed light on how helical zone plates create those beams. The great advantage of this method lays in the well-defined topology, which allows for careful controlling of the phase gradient, and as such the topological charge. We will also discuss the limitations, the stringent need for coherent and astigmatic illumination and the break-down of vortices with high topological charges in many smaller sub-vortices.

In the second part, we delve into recent findings on how OAM beams interact with chiral molecules and magnetic nanostructures, revealing helical dichroism effects that are sensitive to the handedness of the structures. Especially for the realm of naturally chiral molecules, this method is a valuable alternative to the rotation of the polarization plane or the differential absorption of circularly polarized light in the visible light domain. Additionally, we demonstrate how these beams enhance the resolution of high-precision imaging techniques such as ptychography. Potential future applications in the X-ray domain lay in the observation of 3-dimensional magnetic structures –for instance Hopfions –or in super-resolution fluorescence microscopy, dynamic observation of chiral entities, such as Skyrmions, as well as in more systematic investigation of chiral molecules, crystals and metamaterials.

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Ultrafast molecular chirality: a topological connection

Author: Olga Smirnova^{None}

Corresponding Author: olga.smirnova@mbi-berlin.de

An object is said to be chiral if it cannot be superimposed on its mirror image by any rotation. The two mirror images of the same chiral molecule are called enantiomers and are often referred to as “left”- and “right”-handed. While the physical properties of the two enantiomers of the same chiral molecule are nearly identical, the geometric property of chirality leads to vastly different chemical properties of the two enantiomers. The need for rigorous selection of a specific enantiomer, a now standard requirement in drug design, is one of the many reasons behind the ever-greater need for improving sensitivity of chiral sensing.

Yet, standard optical methods of chiral detection still use the same principles as the method discovered by Louis Pasteur in the XIX century: the linear interaction between chiral molecules and light, which becomes chiral-sensitive due to the magnetic field component of the light wave.

Ultrafast non-linear spectroscopies promise to increase the enantio-sensitive signal by three orders of magnitude [1] by removing the need to rely on the interaction with the magnetic field component of light. The second important feature of non-linear light-matter interactions is the opportunity to imprint topological properties of light on matter, presenting an opportunity to achieve topologically robust enantio-sensitive observables.

I will describe our very recent results [2,3] on marrying chiral and topological properties in ultrafast electronic response of chiral molecules in the gas phase, enabling highly efficient and robust chiral observables. I will present three vignettes where topological connection appears in optical or electronic chiral response:

- (i) Chiral topological light: a new concept enabling chiral-sensitive and topologically robust properties of high harmonic emission, generated by such light in chiral molecular gases [2]
- (ii) Geometry of temporal chiral structures: a concept encompassing the emergence of geometric fields in electronic response of chiral molecules [3]
- (iii) Enantio-sensitive exceptional points: chiral topology in non-Hermitian chiral systems [4]

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Thirty-five years of trying –why are we still „developing“ chiral spectroscopy with ultrafast lasers?

Author: Jan Helbing¹

¹ *University of Zurich*

Corresponding Author: jan.helbing@chem.uzh.ch

Since the pioneering transient circular dichroism (CD) experiments of Xie and Simon¹ and Kliger² more than three decades ago, laser technology has developed enormously, and highly stable, ultrashort broad-band pulses for extremely sensitive differential absorption measurements at wavelengths from the mid-IR to the far UV have become routinely available. Yet, progress in time-resolved CD has been much more difficult and slower, and laser-based measurements are still less sensitive and reliable than conventional CD spectrometers, which use much weaker incoherent light sources such as lamps or glow bars.

In the first part of my presentation I will provide an overview of our attempts to boost the sensitivity of laser-based CD detection in the mid-IR and the UV, both via signal enhancement and artifact mitigation^{3–5}, in order to illustrate the difficulty of transferring the measurement principles of non-linear spectroscopy to chirality detection. Artefact reduction in particular requires significantly better laser stability and longer averaging to reach the signal to noise levels of today’s achiral transient absorption measurements⁶.

Another limitation facing transient CD-experiments with ultrashort lasers is the short time-window they can usually cover. Photo-induced changes in molecular reactions often occur on a large range of timescales. While electronic and vibrational relaxation is usually ultrafast, subsequent biomolecular catalytic processes or conformational changes in biomolecules often require hundreds of nanoseconds or microseconds. For example, we seek direct information on local transition dipole couplings in model peptides⁷ by combining transient UV-circular dichroism and site-specific labelling. We will discuss how to couple two femtosecond laser systems to detect absorption and chirality changes under identical excitation and probing conditions over all relevant timescales⁸.

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Ultrafast chiral spectroscopy for stereocontrolled photochemistry

Author: Malte Oppermann¹

¹ *University of Basel, Department of Chemistry*

Corresponding Author: malte.oppermann@unibas.ch

The incorporation of chiral structures into photochemical systems is a powerful strategy to control their functions [1]. For example, uni-directional molecular motors, chiral photocatalysts, and chiral metal nanostructures have achieved exceptional levels of stereocontrol over mechanical motion, energy transfer, and electric charge-carriers on the nanoscale. However, the direct characterization of the underlying chiral excited states remains a formidable experimental challenge, due to a lack of analytical techniques that combine high chiral sensitivity in solution phase with ultrafast time resolution [2].

To address this challenge, we have developed an ultrafast electronic circular dichroism set up that measures the absorption difference of left- and right-circularly polarized laser pulses in photoexcited chiral molecules [3]. Through an ultra-sensitive broadband detection scheme, we are now able to capture ultrafast changes in molecular chirality and follow the stereochemical evolution of the associated photoexcited states with sub-picosecond time resolution. This development has extended the scope of ultrafast chiral spectroscopy to new classes of chiral photochemical phenomena, which I will illustrate via two examples: 1) the identification and control of a new reaction coordinate in the spin-crossover dynamics of Fe(II) complexes [4], and 2) the investigation of the energy and chirality transfer dynamics in a chiral lanthanide-based OLED complex with circularly polarized luminescence (CPL) [5].

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Advances in time-resolved photoelectron circular dichroism of chiral molecules in the gas phase

Author: Vincent Wanie¹

¹ *Deutsches Elektronen-Synchrotron DESY*

Corresponding Author: vincent.wanie@desy.de

PhotoElectron Circular Dichroism (PECD) is an effect driven by electric dipole interactions between an ensemble of chiral molecules and ionizing circularly polarized light [1,2]. It results in a preferential emission direction of photoelectrons along the light propagation axis, leading to an asymmetry that can be observed by recording their angular distribution with an electron detector. Established as a benchmark method to study the static properties of chiral molecules in the gas phase with high sensitivity [3,4], it was extended to the time domain for the investigation of chiral dynamics less than a decade ago [5].

After reviewing the achievements of time-resolved PECD in molecular spectroscopy, I will present the capabilities of our few-femtosecond UV light sources [6,7] to reveal the contribution of ultrafast electron dynamics to the chiral properties of photoexcited neutral molecules [8]. Our results show an ultrafast inversion of the molecular chiroptical response in less than 10 fs after UV excitation, which is captured by modulations of photoelectron angular distributions produced by a time-delayed, circularly polarized ionizing pulse with few-optical-cycle duration. Our theoretical modelling shows that the electronic coherences induced by the UV radiation also generate chiral currents along the molecular structure, offering important prospects for the enantiosensitive, charge-driven reactivity

of chiral molecules [9]. As a proof of principle, I will present a case where chiral electron currents can be used as a tool to select the photodissociation direction of chiral molecules with few-fs resolution. Finally, some concluding remarks will be made on the new opportunities made possible by few-femtosecond PECD.

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Nanoscale structural dynamics by EUV transient gratings

Authors: Filippo Bencivenga¹; Laura Foglia^{None}; Riccardo Mincigrucci²; Claudio Masciovecchio²

¹ *Elettra-Sincrotrone Trieste*

² *Elettra Sincrotrone Trieste*

Corresponding Authors: claudio.masciovecchio@elettra.eu, riccardo.mincigrucci@elettra.eu, laura.foglia@elettra.eu, filippo.bencivenga@elettra.eu

Collective dynamics at the nanoscale in condensed matter is important for advancing both fundamental science and modern technology. The study of heat transport processes, vibrational modes or magnetization dynamics in the sub-100 nm length-scales can greatly benefit from the development of experimental tools for probing such dynamics and on the relevant timescale (i.e. ps and sub-ps) without relying on ad hoc sample's nanostructuration.

In this contribution we present a “contact-less” approach, where the sensitivity to the sub-100 nm length-scale is granted by the use of extreme ultraviolet transient gratings (EUV TG) [1]. We will discuss the application of this new experimental tool (available at the FERMI free electron laser facility in Trieste; Italy) for the study of non-diffusive nanoscale thermal transport in thin membranes of crystalline silicon and amorphous silicon nitride [1,2]. We will also show the possibility to use EUV TG for generating and detecting the dynamics of bulk and surface phonons on a previously inaccessible wavelength range [3], as well as nanoscale magnetization gratings [4] and coherent magnons [5].

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Time-resolved circular dichroism : what can we learn on conformational changes ?

Author: Francois Hache¹

¹ *Laboratoire d'Optique et Biosciences*

Corresponding Author: francois.hache@polytechnique.edu

Time-resolved circular dichroism (CD) is a powerful technique to investigate the dynamics of conformational changes in molecules and in biomolecules. Starting from a pump-probe configuration, it consists in measuring the CD of the pump-excited molecules to gain information on the relevant timescales. Complementary experiments on short (picoseconds) or longer (microseconds) timescales have been set-up based on various polarization-sensitive measurements. However, in order for this technique to be quantitative, it must rely on *a priori* knowledge of the CD spectra before and after photoexcitation or on some simple models. In this talk, such issues will be discussed for several experimental studies : ultrarapid conformational changes in Binaphthol molecules or in the chromophore of Photoactive Yellow Protein on the one hand ; determination of thermodynamic and kinetic parameters in denaturation of poly(Glutamic acid) and DNA G-quadruplexes on the other hand.*emphasized text*