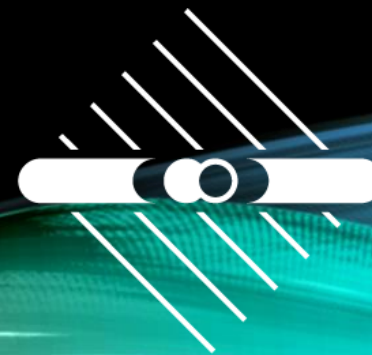


# SOFT MATTER meets ULTRAFAST SPECTROSCOPY SMUS2020

Online Workshop

31<sup>st</sup> August – 1<sup>st</sup> September 2020

MAX PLANCK  
GESELLSCHAFT



cost  
EUROPEAN COOPERATION  
IN SCIENCE & TECHNOLOGY



MAX-PLANCK-INSTITUT  
FÜR KERNPHYSIK

AttoChem  
Attosecond Chemistry

# GOALS

- Gather international scientists from complementary disciplines with the aim to achieve a deeper understanding of the electronic and nuclear dynamics occurring at an extremely wide temporal and spatial scale in LCs, and more generally in soft matter from both experimental and theoretical point of view.
- Present broad overview about current hot topics in research activities of soft matter thus liquid crystals, ultrafast spectroscopy techniques such as THz, transient absorption or high harmonic generation spectroscopy, and computational tools to capture such dynamics.
- Formation of an initial network which will prepare the ground for future joint research activities and establish new collaborations among scientists working in liquid crystals (soft matter) and ultrafast dynamics from both theoretical and experimental point of view.
- Exchange of students/ early career scientists (PhD and postdocs) allowing knowledge transfer across collaborating research groups.

# SESSIONS

- **Current research on Liquid Crystal & Soft Matter: Latest frontiers on material design and theory**
- **Ultrafast dynamics in condensed matter and large molecular systems: preparing the ground for Liquid Crystal ultrafast investigation**
- **AttoChem WG3 session: Towards Attosecond chemistry of Liquid Crystal**

# INVITED PEOPLE <sup>4</sup>

Invited speaker	Research activity
<b>Prof. Paul Kouwer:</b>	Synthesis of soft matter based materials and their structural properties such as hydrogels and different types of liquid crystals.
<b>Prof. Miha Ravnik:</b>	Modeling and simulation of soft and partially ordered matter from macroscopic phenomenology to microscopic molecular modeling.
<b>Prof. Marco Mazza:</b>	Modelling and simulation of nonequilibrium systems: from microorganisms forming biofilms, or flocks of birds self-organizing, to nonequilibrium transitions in granular material.
<b>Prof. Steven Johnson:</b>	Nonlinear dynamics of the atomic structure in solid-state materials.
<b>Prof. Francesca Calegari:</b>	Attosecond ultrafast dynamics in systems with increasing complexity from simple molecules to molecules of biological interest and nanostructured materials.
<b>Dr. Christian Ott:</b>	Attosecond strong field physics in atomic and molecular systems.
<b>Prof. Milutin Kovacev:</b>	High-level research on pulsed lasers and their application from the shortest pulses with single cycle durations via high repetition rate high-energy sources to terawatt peak powers.
<b>Prof. Fernando Martin:</b>	Theoretical study of electron and nuclear dynamics in isolated quantum systems, from atoms to extended systems, interacting with ultrashort laser pulses.
<b>Prof. Angel Rubio:</b>	Modeling and theory of electronic and structural properties of condensed matter.
<b>Prof. Herma Kuppen:</b>	Computational chemistry in organic crystals.
<b>Dr. Hanieh Fattahi:</b>	Femtosecond fieldoscopy.

# PROGRAM

5

co-organised with COST Action CA18222 in Attosecond Chemistry (AttoChem)

	Day 1_Monday August 31 <sup>st</sup>	Day 2_Tuesday September 1 <sup>st</sup>
9:00 <b>start</b>		
9:15	Introduction day 1 - Cattaneo	Introduction day 2 - Cattaneo
9:45	<b>Kower:</b> Challenges in soft matter materials: from responsive to autonomous	<b>Martin:</b> Imaging and controlling electron molecular dynamics: towards attochemistry
10:15	<b>Ravnik:</b> Modelling of flow of light in nematic birefringent structures	<b>Cuppen:</b> To jump or not to jump that is the question
10:45	<b>Johnson:</b> Linear and nonlinear THz spectroscopy of solids	<b>Fattahi:</b> Near-Infrared Molecular Fieldoscopy
11:00	PAUSE	PAUSE
11:30	<b>Rubio:</b> QED-materials and Floquet engineering	<b>Mazza:</b> Liquid crystals out of equilibrium
12:00	<b>Calegari:</b> Tracking electron dynamics in bio-chemically relevant molecules	<b>Kovacev:</b> High-Harmonic Generation from liquid water droplets
12:15	PAUSE	PAUSE
12:45	<b>Ott:</b> The strong-field-driven dipole response in complex systems: Resolving laser-driven femtosecond and attosecond coherent dynamics	<b>Scholz:</b> Ultrafast molecular orbital tomography of a pentacene thin film using time-resolved momentum microscopy at a free-electron laser
13:00	Conclusive thoughts - Cattaneo	Conclusive thoughts - Cattaneo

**Sajadi:** Molecular Liquids under Intense Single Cycle THz Pulses



# TITLES AND ABSTRACTS

# PAUL KOUWER

## **Challenges in soft matter materials: from responsive to autonomous**

Materials in biology are not static but show a large temporal and also spatial variation. For instance, the cells continuously change their microenvironment by depositing and degrading components in their surrounding matrix, thereby always controlling the properties.

Currently in synthetic materials, there is a strong drive towards responsive materials that change their properties after the application of an (external) stimulus. Can we take the next step to generate materials that change properties after a cellular cue or even autonomously?

# MIHA RAVNIK

## **Modelling of flow of light in nematic birefringent structures**

Complex nematic fluids allow for creation or self-assembly of various birefringent structures, which can be used for micro manipulate the flow of light. Here, we show selected examples of modelling the flow of light in selected nematic birefringent structures, including their performance as cloaking devices, photonic crystals, lasers, and metamaterials. Methodologically, we use numerical modelling based on custom adapted solving of Maxwell's equations by (i) Finite Difference Time Domain (FDTD) simulations which can access dynamic fast scales, and (ii) Finite Difference Frequency Domain (FDFD) simulations which solve the light modes emergent in the birefringent profiles. More generally, the aim of this talk is to demonstrate the possible approaches to use birefringent nematic materials in beyond-the-display applications, crucially using different – orders of magnitude shorter- length and time scales.



# STEVE JOHNSON

## **Linear and nonlinear THz spectroscopy of solids**

I give a short overview of the current state of the art in THz spectroscopy of solids, with an emphasis on nonlinear optical effects in the THz regime and how they can be detected. I give some recent examples in correlated electron systems and discuss their possible wider applications in soft matter.

# ANGEL RUBIO

**QED-materials and Floquet engineering**

# FRANCESCA CALEGARI

## **Tracking electron dynamics in bio-chemically relevant molecules**

Attosecond science is nowadays a well-established research field, which offers formidable tools for the investigation and control of electronic processes. The possibility to study molecules of increasing complexity with attosecond time resolution paves the way to disclosing the role of the electron dynamics in the photo-chemistry and photo-biology of complex systems. In this context, I will provide a few examples of the observation of pure electron dynamics in biomolecular building blocks including amino acids and DNA nucleobases. I will then discuss possible approaches for the investigation of these complex molecules in their natural environment, i.e. in water.

# CHRISTIAN OTT

## **The strong-field-driven dipole response in complex systems: Resolving laser-driven femtosecond and attosecond coherent dynamics**

We use ultrashort XUV and few-cycle VIS/NIR optical pulses to probe the absorption spectra of moderately dense gas- and liquid-phase targets, respectively, in Fraunhofer-type transmission geometry. In the time domain, the ultrashort probing pulse triggers a dipole response that then further evolves in a field-free environment. It is this dipole response, emitted in forward direction, that interferes with the incoming pulse and gives rise to sharp spectral signatures in the measured absorption spectrum (i.e., spectral lines), despite extremely short pulses with much larger Fourier spectral bandwidth are being used. Benefitting from the use of time-synchronized ultrashort laser pulses it is possible to further modify this dipole response of the probe-excited system with a moderately strong laser field. Coherent control of the absorption line shapes can thus be achieved with access to both amplitude and phase, and the laser imprint on the time-dependent dipole response can be directly extracted from the measured spectra in real time, with sub-femtosecond temporal resolution. In this talk I will present experimental results on the real-time buildup dynamics of complex spectral structures encoded in the Rydberg series of overlapping autoionizing Fano resonances in helium. Furthermore, I will discuss first experimental results on a much more complex target, aluminum phthalocyanine chloride dissolved in ethanol, as an example for the coherent strong-field control of liquid-phase VIS absorption spectra.

# FERNANDO MARTIN

## Imaging and controlling electron molecular dynamics: towards attochemistry

Sudden excitation or ionization of a molecule by visible, UV or XUV light is usually followed by charge redistribution on a time scale ranging from hundreds attoseconds to a few-femtoseconds, which can be followed by fragmentation of the remaining molecular cation up to a few picoseconds. Such complex dynamics arises from the coherent superposition of electronic states covered by the broadband pulse and from rearrangements in the electronic structure of the molecular cation due to electron correlation. To investigate these ultrafast processes, attosecond pump-probe spectroscopies have been shown to be very valuable tools [1-9]. In this talk I will present the results of gas-phase molecular pump-probe experiments and theoretical simulations in which several molecules and solids are ionized and subsequently probed in order to obtain the complete electronic movie. I hope that these results will help to guide future experimental efforts in more complicated molecules, even in the liquid phase, and will open the door to the control of charge transfer [10]. These efforts should be accompanied by the development of new theoretical methods able to accurately describe the ionization continuum of molecules in the gas [11] and condensed phases.

### References

- [1] G. Sansone et al, *Nature* **465**, 763 (2010).
- [2] F. Calegari et al, *Science* **346**, 336 (2014).
- [3] A. Palacios, A. González-Castrillo and F. Martín, *Proc. Natl. Acad. Sci.* **111**, 3973 (2014).
- [4] P. Ranitovic et al, *Proc. Natl. Acad. Sci.* **111**, 912 (2014).
- [5] A. Trabattoni et al, *Phys. Rev. X* **5**, 041053 (2015).
- [6] M. Lara-Astiaso et al, *Faraday Discussions* **194**, 41 (2016).
- [7] M. Lara-Astiaso et al, *J. Phys. Chem. Lett.* **9**, 4570 (2018).
- [8] L. Cattaneo et al, *Nature Physics* **14**, 733 (2018).
- [9] E. Plésiat, M. Lara-Astiaso, P. Decleva, A. Palacios, and F. Martín, *Chemistry: Eur. J.* **24**, 12061 (2018).
- [10] M. Nisoli, P. Decleva, F. Calegari, A. Palacios, and F. Martín, *Chem. Rev.* **117**, 10760 (2017).
- [11] A. Palacios and F. Martín, *WIREs Comp. Mol. Sci.*, e1430 (2020).

# HERMA CUPPEN

## **To jump or not to jump that is the question**

Mechanically responsive molecular crystals that reversibly change shape in response to external stimuli are a promising class of materials. Particularly interesting are thermosalient crystals, which are crystals that jump or leap upon heating or cooling due to a thermally induced single-crystal-to-single-crystal (SCSC) phase transition. Several cases of jumping crystals have been reported in the literature, but a mechanistic understanding of the underlying phase transition is lacking. I will show how a combination of experimental and simulation techniques can lead to a better understanding of the underlying mechanism of these transitions.

# HANIEH FATTAHI

## Near-Infrared Molecular Fieldoscopy

Linear spectroscopy in near-infrared spectral range provides information on the molecular composition, structure, and conformation, affording tremendous potential for high-resolution, in-depth, label-free biological spectromicroscopy [1]. Broadband measurements in this spectral region have been carried out mainly in the frequency domain, which suffers from two limitations: i) the detection of small absorption differences is limited to the intensity noise of the source, and ii) the low detection dynamic range.

To mitigate above-mentioned limitations, we introduce the concept of molecular Fieldoscopy. In molecular Fieldoscopy, an ultrashort, phase-coherent pulse is used for molecular excitation and afterwards the transmitted complex electric field that contains the molecular response is measured directly. Here, the excitation pulse is confined to a time window of tens of femtoseconds, which is significantly shorter than molecular response. Therefore, the molecular response is detected free of background and the effect of the intensity noise of the source is eliminated. Furthermore, measuring the complex electric field allows for extracting the full spectral phase information of the molecular response, adding a new dimension to the gained spectroscopic data.

1 F. F. Jöbsis, "Noninvasive, infrared monitoring of cerebral and myocardial oxygen sufficiency and circulatory parameters." *Science* 198, 1264–1267 (1977).

# MARCO MAZZA

## **Liquid crystals out of equilibrium**

I will discuss some recent results on liquid crystals in nonequilibrium. First, hydrodynamic cavitation, the nucleation of vapour in liquids. With a combination of microfluidic experiments, nonequilibrium molecular dynamics simulations and theoretical arguments, we have studied flow-induced cavitation in a nematic fluid.

The cavitation domain nucleates due to sudden pressure drop upon flow past a cylindrical obstacle within a microchannel. For an anisotropic fluid, the inception and growth of the cavitation domain ensued in the Stokes regime, while no cavitation was observed in isotropic liquids flowing under similar hydrodynamic parameters.

Second, some ongoing work on the dynamics of active nematic droplets will be briefly discussed.



# MILUTIN KOVACEV

## **High-Harmonic Generation from liquid water droplets**

An overview will be given on the subject of strong-field interactions with liquid water droplets. We will present our experimental measurements and theoretical simulations. The talk will also address the historical development, current challenges and future perspectives.

# MARKUS SCHOLZ

## **Ultrafast molecular orbital tomography of a pentacene thin film using time-resolved momentum microscopy at a free-electron laser**

Markus Scholz<sup>1</sup>, Kiana Baumgärtner<sup>2</sup>, Christian Metzger<sup>2</sup>, Manuel Izquierdo<sup>1</sup>, Dmytro Kutnyakhov<sup>3</sup>, Serguei Molodtsov<sup>1</sup>, Friedrich Reinert<sup>2</sup>, Anders Madsen<sup>1</sup>

<sup>1</sup>European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany, Markus.Scholz@xfel.eu

<sup>2</sup>Experimentelle Physik 7, Julius-Maximilians-Universität, Am Hubland, 97074 Würzburg, Germany

<sup>3</sup>Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22607 Hamburg, Germany

In the last decade, orbital tomography has emerged as an exciting extension of the photoemission technique for imaging the wave functions of molecules in thin films. Recently, we pioneer time-resolved orbital tomography of a pentacene film using a free-electron laser (FEL).

Understanding and control of photon-induced dynamics of molecules on solid surfaces are of essential importance for surface chemistry as well as for design of novel devices. Time-resolved tomography of molecular wave functions will provide unprecedented insight into photon-induced dynamics and chemical reactions. This is of paramount importance for designing and optimizing functional molecular materials and their optoelectronic properties.

Using condensed bi-layers of pentacene atop Ag(110) as example, we demonstrate that time-resolved orbital tomography is feasible using a FEL. The high repetition rate of the FLASH FEL allowed us to capture photoelectron momentum maps of molecular valence orbitals with a time-resolution of a few hundred femtoseconds. In pump-probe experiments we observed a transient signal 400 meV above the Fermi edge. The real-space reconstruction of this feature aligns with the in-plane ordering of pentacene on Ag(001). The wave function expands over two adjacent molecules located at the pentacene-Ag interface suggesting a novel delocalized electronic state.

# MOHSEN SAJADI

## Molecular Liquids under Intense Single Cycle THz Pulses

Mohsen Sajadi<sup>1,2</sup>

<sup>1</sup>*Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany.*

<sup>2</sup>*Department of Chemistry, University of Paderborn, Paderborn, Germany.*

Intense single-cycle terahertz (THz) pulses have great potential to reveal detailed insight into the structural dynamics of molecular systems.<sup>1,2</sup> Their strong coupling to the low-frequency electric and nuclear degrees of freedom determines the intermolecular energy potential surface and reveals the energy dissipation paths in liquids.

The intense THz magnetic fields are used to induce ultrafast rotation of the plane of polarization of a short optical pulse traversing polarizable molecular liquids. We explain the effect as the bending of the electric polarization induced by the optical short laser pulse under the influence of a Lorentz force emanated from the THz magnetic field. The observed effect scales linearly with the THz magnetic field and quadratically with the molecular polarizability.<sup>3</sup>

The intense THz electric fields are used to show the tunability of the molecular rotational distributions in liquids, by creating partially aligned and anti-aligned distributions. The mechanism of this process is discussed in terms of an efficient mechanical THz torque to the permanent molecular dipoles. It is shown that by tuning the THz pulse frequency, phase and also the strength of the THz electric coupling to the molecular dipole moment, different rotational distributions can be achieved. THz-control of the molecular rotational distribution in liquids opens a new path to study intermolecular interactions and offers a new means for coherent control of chemical reactions.<sup>4</sup>

### References

1. T. Kampfrath, M. Wolf, M. Sajadi, *J. Phys. Chem. Lett.* 9 (2018).
2. M. Sajadi, M. Wolf, T. Kampfrath, *Nat. Comm.* 8 (2017).
3. V. Balos, G. Bierhance, M. Wolf, M. Sajadi, *Phys. Rev. Lett.* 124, (2020).
4. H. Elgabarty, et al., *Sci. Adv.* eaay7074 (2020).

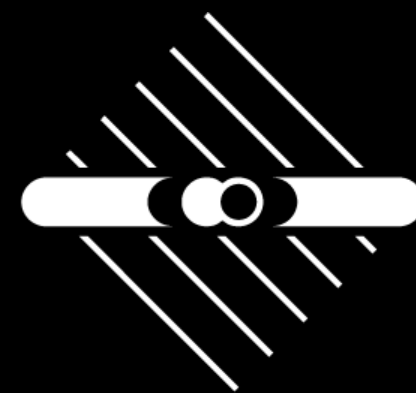
# AFFILIATIONS

- Prof. Miha Ravnik: University of Ljubljana (SL)
- Prof. Marco Mazza: Loughborough University (UK)
- Prof. Herma Kuppen: Radboud University (Nijmegen, NH)
- Prof. Fernando Martin: Universidad Autónoma de Madrid (SP)
- Prof. Angel Rubio: Max Planck Institute for the Structure and Dynamics of Matter (Hamburg, DE)
- Prof. Francesca Calegari: DESY (Hamburg, DE)
- Prof. Paul Kower: Radboud University (Nijmegen, NH)
- Prof. Steven Johnson: ETH Zurich (CH)
- Prof. Milutin Kovacev: Leibniz Universität (Hannover, DE)
- Dr. Hanieh Fattahi: Max Planck Institute for the Science of Light (Erlangen, DE)
- Dr. Christian Ott: Max Planck Institute for Nuclear Physics (Heidelberg, DE)
- Dr. Markus Scholz: European XFEL (Schenefeld, DE)
- Dr. Moshen Sajadi: Fritz-Haber-Institut der Max-Planck-Gesellschaft (Berlin, DE)

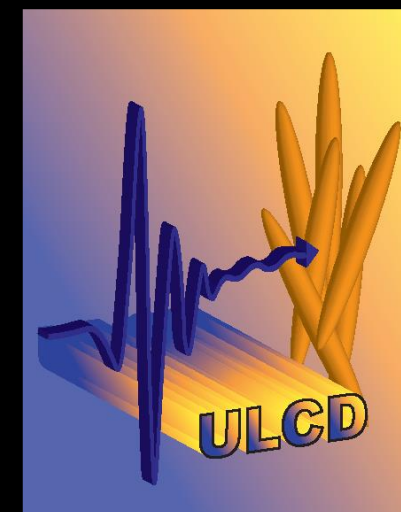
# EMAILS

- [p.kouwer@science.ru.nl](mailto:p.kouwer@science.ru.nl)
- [angel.rubio@mpsd.mpg.de](mailto:angel.rubio@mpsd.mpg.de)
- [johnson@phys.ethz.ch](mailto:johnson@phys.ethz.ch)
- [francesca.calegari@desy.de](mailto:francesca.calegari@desy.de)
- [M.G.Mazza@lboro.ac.uk](mailto:M.G.Mazza@lboro.ac.uk)
- [fernando.martin@uam.es](mailto:fernando.martin@uam.es)
- [christian.ott@mpi-hd.mpg.de](mailto:christian.ott@mpi-hd.mpg.de)
- [miha.ravnik@fmf.uni-lj.si](mailto:miha.ravnik@fmf.uni-lj.si)
- [H.Cuppen@science.ru.nl](mailto:H.Cuppen@science.ru.nl)
- [kovacev@iqo.uni-hannover.de](mailto:kovacev@iqo.uni-hannover.de)
- [hanieh.fattahi@mpl.mpg.de](mailto:hanieh.fattahi@mpl.mpg.de)
- [markus.scholz@xfel.eu](mailto:markus.scholz@xfel.eu)
- [sajadi@fhi-berlin.mpg.de](mailto:sajadi@fhi-berlin.mpg.de)
- [cattaneo@mpi-hd.mpg.de](mailto:cattaneo@mpi-hd.mpg.de)

**MAX PLANCK**  
GESELLSCHAFT



MAX-PLANCK-INSTITUT  
FÜR KERNPHYSIK



**cost**  
EUROPEAN COOPERATION  
IN SCIENCE & TECHNOLOGY



**AttoChem**  
Attosecond Chemistry

“This activity is co-organized by COST through CA18222 Action funds. COST is supported by the EU Framework Programme Horizon 2020. More information at <https://www.cost.eu/actions/CA18222>”