Geneva chemistry & biochemistry days 2021

TH 14 January 2021, 09:00–16:20
FR 15 January 2021, 09:00–11:50
LIVE STREAMING VIRTUAL EVENT
https://bit.ly/2KTSpEU
No registration required

Prof. Yves Barral
Eidgenössische Technische Hochschule Zürich

Prof. Jonathan Clayden
University of Bristol

Dr Ivano Tavernelli
IBM Research Zürich

Prof. Oliver Wenger
Universität Basel

Junior Speakers:

Jafar Afshani Aghajari • Mireia Andreu-Carbó • Ryo Asakura • Ewa Banach • Lluc Farrera Soler • Michele Garbo • Daniel Hummel • Pitchnaree Kraikaew • Canwei Mao • Rémi Martinent • Manon Mottet • Johann Nuck • Pavol Ondrisek • Rémi Patouret • Anh-Tuan Pham • Niccolo Ricardi • Yanan Wang •

FACULTÉ DES SCIENCES
SECTION DE CHIMIE ET BIOCHIMIE

UNIVERSITÉ DE GENÈVE

FOREWORD

The *Section de chimie et biochimie*, University of Geneva, has the pleasure to announce the 11th edition of its "***Geneva Chemistry & Biochemistry Days***". Due to the sanitary situation, the event will be held at distance, via the Zoom interface (https://unige.zoom.us/j/92682442652 + code GCBD2021 or directly **https://bit.ly/2KTSpEU**; see tips on page iii), but we are convinced that the spirit of the event will not be impaired by this mishap.

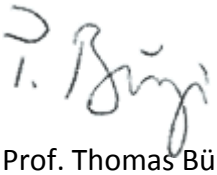
The vocation of the event is to give our students who are close to finishing their PhD studies the opportunity to present their research as attractive speed talks to an audience from academia and industry, and the steering committee is glad to welcome you in this context.

Four distinguished lecturers further enrich the programme. Our four departments have invited them, and they will illustrate the extent and the quality of top-level fundamental research in chemistry and biochemistry today.

Our BSc and MSc students are welcome to smell the very flavour of the research held in our School and abroad, and to learn a bit more about how to present punchy results to a scientific audience.

We expect that the event will catalyse fruitful discussions between young and advanced researchers, and give our students an opportunity to get ready for their professional career, yet offering our guests an overview of the quality of the fundamental research performed in our School.

Looking forward to welcoming you at this event, we hope that you will enjoy the lectures and interactions!



Prof. Thomas Bürgi
Président de la Section de chimie et biochimie

Steering and organising committee

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PROGRAMME – THURSDAY, 14 JANUARY https://bit.ly/2KTSpEU

Chair: **Prof. Karsten Kruse** (Senior Speaker)
Dr Huayan Yang (Junior Speakers)

09:00--09:05	**Prof. Thomas Bürgi**	Welcome message
09:05--09:55	**Prof. Yves Barral** Eidgenössische Technische Hochschule Zürich	Ageing: Insights from a good old fungal friend
09:55--10:10	**Lluc Farrera Soler**	Immunological epitope mapping of the SARS-CoV-2 spike protein
10:10--10:25	**Jafar Afshani Aghajari**	Synthesis and luminescence properties of strontium aluminate nanospheres as long persistent phosphors
10:25--10:40	**Mireia Andreu-Carbó**	Walking kinesins stabilize their microtubule tracks along the shaft
10:40--11:00	Break	2 Discussion Rooms available from within the main Conference Room
11:00--11:15	**Michele Garbo**	Access to optically active 7-membered rings by a 2-step catalytic sequence
11:15--11:30	**Ewa Banach**	Photoswitchable Au_{25} nanocluster assemblies
11:30--11:45	**Canwei Mao**	Separating boundary potential changes at thin solid contact ion transfer voltammetric membrane electrodes
11:45--12:00	**Rémi Martinent**	Oligomers of cyclic oligochalcogenides for enhanced cellular uptake
12:00--13:30	Lunch break	2 Discussion Rooms available from within the main Conference Room

Chair: **Prof. Stefan Matile** (Senior Speaker)
Dr Luca Barberi (Junior Speakers)

13:30--13:45	**Johann Nuck**	Polydiacetylene-peptide interaction mechanism in mixed lipid systems
13:45--14:00	**Daniel Hummel**	SH3 domains regulate spatio-temporal assembly of actin nucleation promoting factors in endocytosis
14:00--14:15	**Pavol Ondrisek**	Modular synthesis of mono and dicationic [4]helicenes with complementary biaryl atropisomerism
14:15--14:30	**Niccolo Ricardi**	Frozen Density Embedding Theory: What densities can one use? What errors can one expect?
14:30--14:45	Break	2 Discussion Rooms available from within the main Conference Room
14:45--15:00	**Pitchnaree Kraikaew**	Ultra-sensitive pH measurements with a coulometric principle
15:00--15:15	**Rémi Patouret**	Total synthesis of goyazensolide and identification of the first importin-5 inhibitor
15:15--15:30	**Yanan Wang**	Amplification of enantiomeric excess by dynamic inversion of enantiomers in deracemization of Au_{38} clusters
15:30--16:20	**Prof. Jonathan Clayden** University of Bristol	Exploiting molecular conformation for biomimetic function and reactivity
16:20-	Evening break	2 Discussion Rooms available from within the main Conference Room

PROGRAMME – FRIDAY, 15 JANUARY https://bit.ly/2KTSpEU

Chair: **Prof. Claude Piguet** (Senior Speaker)
Dr Elena Zdrachek (Junior Speakers)
Prof. Tomasz Wesolowski (Senior Speaker)

09:00--09:50	**Prof. Oliver Wenger** Universität Basel	Photoactive coordination compounds based on Earth-abundant metals
09:50--10:05	**Manon Mottet**	Single cell analysis unravels a perturbation of the cell cycle of the host *D. discoideum* during infection by *M. marinum*
10:05--10:20	**Anh-Tuan Pham**	Peptide stapling with Anion-π Catalysts
10:20--10:35	Break	2 Discussion Rooms available from within the main Conference Room
10:35--10:50	**Ryo Asakura**	4 V room-temperature all-solid-state sodium battery enabled by a passivating cathode/electrolyte interface
10:50--11:40	**Dr Ivano Tavernelli** IBM Research Zürich	Quantum computing and its applications in chemistry and physics
11:40--11:45	**Prof. Stefan Matile**	Awards for the best oral Junior presentations
11:45--11:50	**Prof. Thomas Bürgi**	Concluding remark

TIPS FOR A SMOOTH ZOOM EXPERIENCE

The event will be held at distance, via the Zoom interface. During the whole event, the main Conference Room will be **https://bit.ly/2KTSpEU**

From the main Conference Room, two parallel Discussion Rooms will be available; these Discussion Rooms can be accessed anytime during the event.

On Wednesday, 13 January, from 8:30 to 17:30, Junior Speakers and Senior Speakers are invited to **test connexion and screen sharing capabilities** in the conference room with the event administrator; please feel free to join!

On Thursday, 14 January, and Friday, 15 January, the Conference Room and the parallel Discussion Rooms will be open to the public from 8:30 to 17:30 (Thursday) and from 8:30 to 13:00 (Friday). Participants are invited to create additional parallel Discussion Rooms for their private use.

You can enter the Conference Room or the Discussion Rooms directly via your browser, but the quality of the broadcast may be limited under certain circumstances; for a better experience, it is recommended to download the autonomous Zoom engine via the Zoom Download Centre at **https://zoom.us/download**.

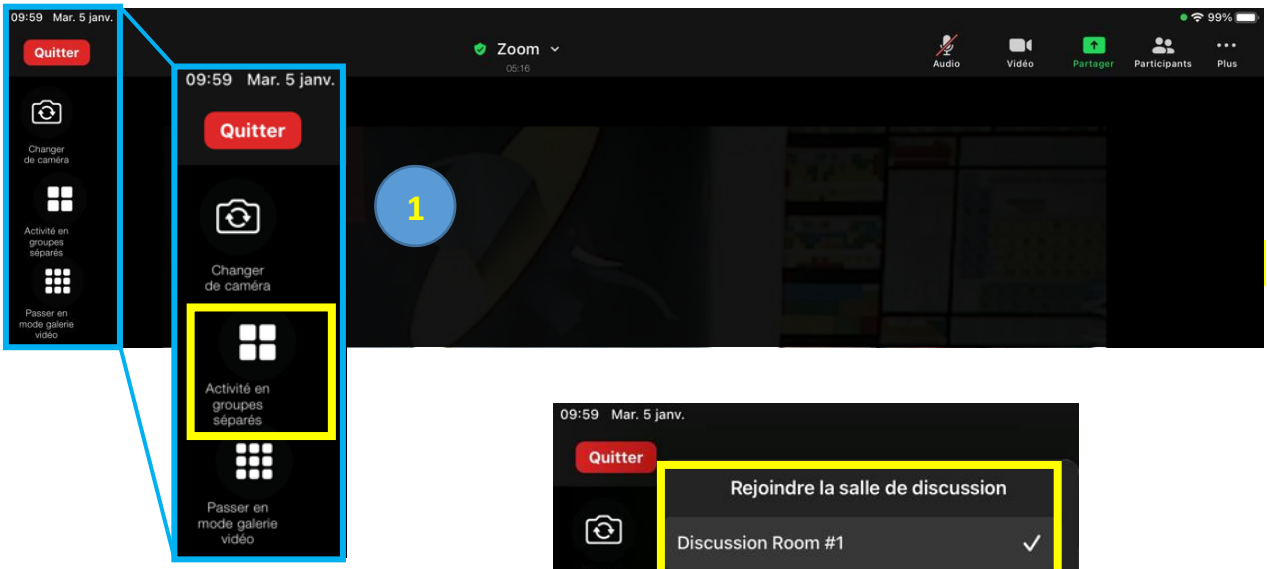
To guarantee a clear voice connexion, it is advised to use an external microphone or headset (*e.g.* USB-connected) instead of the built-in microphone/speaker mounted in your computer.

When reaching the Conference Room as a participant, please make sure that your microphone is switched to OFF. If you have a slow- or medium-speed connexion, you can increase the bandwidth by switching your camera to OFF.

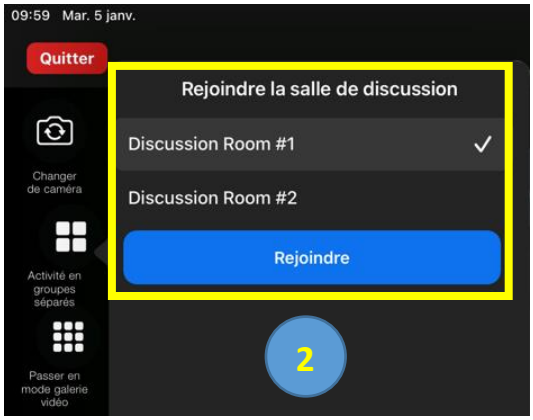
The chairwomen, chairmen, and speakers will be granted administrator rights, to take over the screen-sharing option of Zoom at the beginning of their talk.

To **join a parallel Discussion Room** and then come back to the main Conference Room, proceed as follows (example below is on an iPad-based Zoom session, in French; the views may differ depending on the computer used):

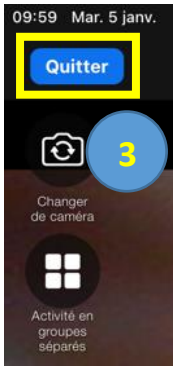
1) Select the bloc of ***Activities in separate groups***:



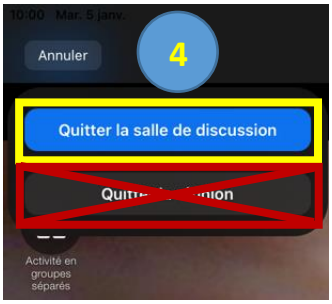
2) Select the required ***Discussion Room*** and click on ***Join***:



3) When finished, click on ***Quit*** to join back the main Conference Room:



4) Select ***Quit the Discussion Room*** (and <u>not</u> *Quit the Meeting*, which would kick you out of the main Conference Room!); you will be automatically brought back to the main Conference Room:



DIRECT ZOOM LINK TO THE CONFERENCE ROOM

https://bit.ly/2KTSpEU or scan the QR code

Ageing: Insights from a good old fungal friend

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Over the years, budding yeast has turned in a powerful system for studying the ageing process at the cellular level. At the same time, since the ageing yeast mother cell generates young daughter cells, budding yeast emerged as a key model system for studying the mechanisms of cellular rejuvenation.

Recently, our laboratory has addressed the role of lateral diffusion barriers that forms in the ER membranes at the mother-bud neck in the retention of ageing factors in the mother cell and the resetting of age in the daughter cells. These studies support the idea that there are mainly two types of ageing factors accumulating in yeast mother cells: protein aggregates and circles of DNA emanating from the genome by homologous recombination. In the last few years, we have been focusing on deciphering the relationships between these ageing factors and the mechanisms by which they cause the fitness decay and the eventual death of ageing cells.

Here, I will present our recent data on how diffusion barriers form and confine age to yeast mother cells, using saturated lipids such as ceramide to assemble filters in the plan of ER membranes. I will also present our most recent data about how nuclear and cytoplasmic ageing (reflected by DNA circles and protein aggregates, respectively) interfere with each other to cause the progressive demise of ageing cells. Particularly, I will provide evidence for successive events of nuclear pore remodelling and discuss how these contribute to the propagation of ageing from the nucleus to the cytoplasm.

Exploiting molecular conformation for biomimetic function and reactivity

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Nature solves many challenges of molecular reactivity and molecular communication by making use of exquisite control of molecular conformation.

The lecture will explore the use of synthetic molecules with well-defined conformations to induce new modes of reactivity of value in synthesis for example, the metal-free arylation and vinylation of enolates[1,2] and amines,[3] or the synthesis of medium rings.[4,5]

It will also explore the ways in which structures with switchable global conformation can be exploited in systems that exhibit artificial signal transduction in the form of remote stereocontrol and in the design and construction of artificial transmembrane receptors.[6,7]

References:

1. Leonard D.J., Ward J.W., Clayden J. *Nature* **2018**, *562*, 105.
2. Abas H., Mas Roselló J., Amer M.M., Durand D.J., Groleau R.R., Fey N., Clayden J. *Angew. Chem. Int. Ed.* **2019**, *58*, 2418.
3. Costil R., Dale H.J.A., Fey N., Whitcombe G., Matlock J.V., Clayden J. *Angew. Chem. Int. Ed.* **2017**, *56*, 12533.
4. Hall J.E., Matlock J.V., Ward J.W., Gray K.V., Clayden J. *Angew. Chem. Int. Ed.* **2016**, *55*, 11153.
5. Costil R., Lefebvre Q., Clayden J. *Angew. Chem. Int. Ed.* **2017**, *56*, 14602.
6. De Poli M., Zawodny W., Quinonero O., Lorch M., Webb S.J., Clayden J. *Science* **2016**, *352*, 575.
7. Lister F.G.A., Le Bailly B.A.F., Webb S.J., Clayden J. *Nature Chem.* **2017**, 9, 420.

Quantum computing and its applications in chemistry and physics

Ivano TAVERNELLI

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Quantum computing is emerging as a new paradigm for the solution of a wide class of problems that are not accessible by conventional high performance classical computers. Quantum computers can in principle efficiently solve problems that require exponential resources on classical hardware, even when using the best-known classical algorithms. In the last few years, several interesting solutions with potential quantum speedup have been brought forward in the domain of quantum physics, like the quantum phase estimation and the hybrid variational quantum eigensolver[1] for the solution of optimization problems.

The original idea that a quantum computer can potentially solve many-body quantum mechanical problems more efficiently than classical computers is due to R. Feynman who proposed the use of quantum algorithms to investigate the fundamental properties of nature at the quantum scale. In particular, the solution of the electronic structure and statistical mechanics problems is a challenging computational task as the number of resources increases exponentially with the number of degrees of freedom. Thanks to the development of new quantum technologies witnessed over the last decades, we have now the possibility to address this class of problems with the help of quantum computers. To achieve this goal, new quantum algorithms able to best exploit the potential quantum speedup of state-of-the-art noisy quantum hardware have also been developed.[2,3]

In this talk, I will first introduce the basics of quantum computing using super-conducting qubits, focusing on those aspects that are crucial for the implementation of quantum chemistry and physics algorithms. In the second part, I will highlight the potential advantages of the new generation of quantum algorithms for applications in electronic structure calculations for ground[4] and excited states,[5] molecular dynamics,[6] and statistical physics.[7]

References:

1. Peruzzo A. et al. *Nature Comm.* **2014**, *5*, 4213.
2. Moll N. et al. *Quantum Sci. Technol.* **2018**, *3*, 030503.
3. Kandala A. et al. *Nature* **2017**, *549*, 242.
4. Baroutsos P. et al. *Phys. Rev. A* **2018**, *98*, 022322.
5. Ganzhorn M. et al. *Phys. Rev. Appl.* **2019**, *11*, 044092; Ollitrault P. et al. *arXiv* **2019**, 1910.12890.
6. Sokolov I.O., Barkoutsos P., Moeller L. et al. *arXiv* **2020**, 2008.08144.
7. Robert A. et al. *arXiv* **2019**, 1908.02163.

Photoactive coordination compounds based on Earth-abundant metals

Oliver WENGER

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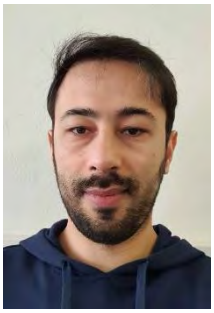
Photochemical reactions and photophysical processes such as luminescence rely on electronically excited states with lifetimes of at least a few nanoseconds. To obtain compounds with such excited-state lifetimes, a very stringent set of criteria must be fulfilled, and this limits the available chemical space for photoactive compounds severely.

In this talk, I will try to explain why precious metals such as ruthenium or iridium have received so much attention in the past, and I will discuss recent advances in the search for photoactive complexes made from cheaper and more abundant metals such as iron, chromium, and molybdenum.

Synthesis and luminescence properties of strontium aluminate nanospheres as long persistent phosphors



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$SrAl_2O_4:Eu^{2+},Dy^{3+}$ is commonly accepted as the best non-radiative long persistent phosphor in terms of luminescence intensity and afterglow duration, known up to date.[1] However, the big particle sizes ranging in between 20-100 µm limits its application predominantly to displays and safety signs. Despite many attempts to develop new synthetic methods, nanosized particles of strontium aluminate with homogeneous morphology have not been obtained yet. Most of the reports on the strontium aluminate nanoparticles have applied top-down method, which results in very bad morphology and wide size distribution.

We have developed a new synthesis method for creating strontium aluminate precursor nanoparticles with spherical morphology and narrow size distribution, which can be used for different applications. Moreover, investigation of the luminescence properties of these nanoparticles doped with Eu^{3+} leads to very interesting results.

Reference:

1. Rocío E.R.H., Fernando R.M., Miguel Á.R., José F.F., *Renew. Sust. Energ. Rev.* **2018**, *81*, 2759.

Walking kinesins stabilize their microtubule tracks along the shaft

Mireia ANDREU-CARBÓ

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Microtubules (MTs) are highly dynamic polymers forming a network within the cell that defines cytosol architecture, shape and polarity of the cell. Recent studies proposed that not only the MT-tip but also the MT-shaft is a dynamic structure that influences MT dynamics.[1-4] These dimer exchanges form GTP-tubulin islands along the shaft and have been shown to be rescue sites[2-4] leading to longer MTs with increased lifetime.[3] MTs also serve as tracks for molecular motors that walk long distances along them to deliver cargoes throughout the cell.

Our *in vitro* results show that running kinesin-1 increases lateral tubulin turnover in a concentration-dependent manner. The kinesin-MT system is very efficient: within 15 min 20 % of the MT shaft is renewed. This rise of GTP-tubulin islands along the shaft impacts on MT dynamics: by increasing rescue frequency MT mass is increased. In order to understand the effect of walking kinesins on MT network in cells we acutely activated native kinesin-1.

Our observations demonstrate that active kinesin-1 not only uses MTs as track but changes its MT dynamic properties by increasing MT rescue frequency, lifetime and, therefore, MT density within minutes. In this context, local walking of kinesin-1 along the shaft modifies its tracks to ensure their stability and, in turn, impacts on the microtubule density and enhances cell polarity.

References:

1. Schaedel L., John K., Gaillard J., Nachury M.V., Blanchoin L., Thery M. *Nat. Mater.* **2015**, *14*, 1156.
2. de Forges H., Pilon A., Cantaloube I., Pallandre A., Haghiri-Gosnet A.M., Perez F., Poüs C. *Curr. Biol*. **2016**, *26*, 3399.
3. Aumeier C., Schaedel L., Gaillard J., John K., Blanchoin L., Théry M. *Cell Biol*. **2016**, *18*, 1054.
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4 V room-temperature all-solid-state sodium battery enabled by a passivating cathode/electrolyte interface

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Room-temperature operation of high-voltage all-solid-state batteries requires solid electrolytes that combine high cation conductivity (≥1 mS cm^{-1}), compatibility with lithium or sodium metal anodes and high-voltage cathodes, and intimate electrode/solid electrolyte interfaces. However, none of the solid electrolytes has fulfilled these material properties so far. Hydroborates, a yet underexplored class of solid electrolytes,[1,2] are highly conductive at room temperature and compatible with lithium and sodium metal anodes. However, the cell voltage was limited up to 3 V in previous reports,[3–6] mainly due to the limited electrochemical oxidative stability of the solid electrolytes.[7]

Here I show that a hydroborate solid electrolyte, consisting of two kinds of hydroborate anions with different oxidative stability limits, can be effectively stabilized in contact with a 4 V-class cathode.[8] At high voltage, the less stable anions tend to form a passivating interphase layer upon electrochemical oxidation at the cathode/solid electrolyte interface, while the more stable anions remain intact in the interphase layer, maintaining cation conduction. The self-passivating interphase enables the first stable cycling of a 4 V-class hydroborate-based all-solid-state battery employing a sodium metal anode and a cobalt-free, high-voltage cathode. The cells exhibit a discharge capacity of 100 mAh g^{-1} at C/5 and an excellent capacity (78%) and energy (76%) retention after >800 cycles at room temperature. Applying external pressure enables a discharge capacity of >110 mAh g^{-1} at C/10 with a high areal capacity close to 1.0 mAh cm^{-2}. This work records the highest discharge cell voltage and specific energy at the active material level among all reported all-solid-state sodium batteries, demonstrating the attractive material properties and potential of hydroborates that surpass intensively investigated oxides and sulfides as solid electrolytes for high-voltage all-solid-state batteries.

References:

1. Duchêne L., Remhof A., Hagemann H., Battaglia C. *Energy Storage Mater.* **2020**, *25*, 782.
2. Brighi M., Murgia F., Černý R. *Cell Rep. Phys. Sci.* **2020**, *1*, 100217.
3. Duchêne L., Kühnel R.-S., Stilp E., Cuervo Reyes E., Remhof A., Hagemann H., Battaglia C. *Energy Environ. Sci.* **2017**, *10*, 2609.
4. Duchêne L., Kim D. H., Song Y. B., Jun S., Moury R., Remhof A., Hagemann H., Jung Y. S., Battaglia C. *Energy Storage Mater.* **2020**, *26*, 543.
5. Murgia F., Brighi M., Černý R. *Electrochem. Commun.* **2019**, *106*, 106534.
6. Kim S., Oguchi H., Toyama N., Sato T., Takagi S., Otomo T., Arunkumar D., Kuwata N., Kawamura J., Orimo S. *Nat. Commun.* **2019**, *10*, 1081.
7. Asakura R., Duchêne L., Kühnel R.-S., Remhof A., Hagemann H., Battaglia C. *ACS Appl. Energy Mater.* **2019**, *2*, 6924.
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Photoswitchable Au_{25} nanocluster assemblies

Ewa BANACH

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Ultrasmall (< 2 nm diameter) ligand-protected gold nanoclusters (Au NCs) are a unique class of materials characterized by their well-defined structure, precise atomic composition, and size-dependent properties that are distinct from their bulk metal counterparts. Moreover, their physicochemical properties change considerably depending on the number of constituent atoms. For these reasons, Au NCs show promise as building blocks for functional nanomaterials, such as optoelectronic devices or sensors.[1] However, for such applications the ability to switch between different states is necessary. Herein, a photoswitchable system based on Au_{25} nanocluster assemblies is presented (Fig.1). We employ a carefully-designed photo-switchable bridging dithiol ligand based on a dithienylethene (DTE) moiety[2] to modulate the electronic communication between linked Au nanoclusters.

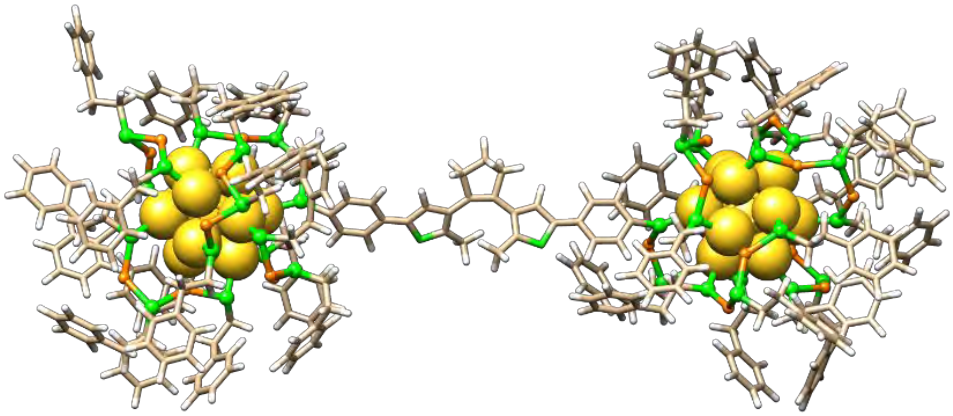

Fig. 1. Dimeric Au_{25} assembly covalently linked *via* switchable photoligand.

The bottom-up formation of nanoassemblies was achieved by reacting archetypical thiolate-protected $Au_{25}(SCH_2CH_2Ph)_{18}$ clusters[3] with the photoligand in solution *via* ligand-exchange. The linked Au_{25} species were characterized with SAXS and MALDI-MS to identify their size. Importantly, reversible photoisomerization of the DTE unit in the bridging ligand was found to induce significant changes in the photophysical behaviour of the formed Au_{25} multimers compared to monomeric Au_{25} nanoclusters. In this presentation, the outcomes of spectroscopic studies combined with DFT calculations will be briefly discussed to offer insight into the complicated processes governing the light-induced physicochemical changes occurring in the formed Au_{25} multimers.

References:

1. Jin R., Zeng Ch., Zhou M., Chou Y., *Chem. Rev.* **2016**, *116*, 10346.
2. Irie M., *Chem. Rev.* **2000**, *100*, 1685.
3. Kang X., Chong H., Zhu M. *Nanoscale* **2018**, *10*, 10758.

Immunological epitope mapping of the SARS-CoV-2 spike protein

Lluc FARRERA SOLER

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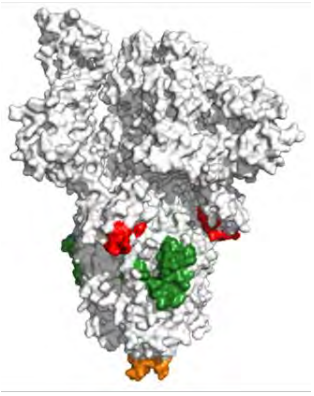
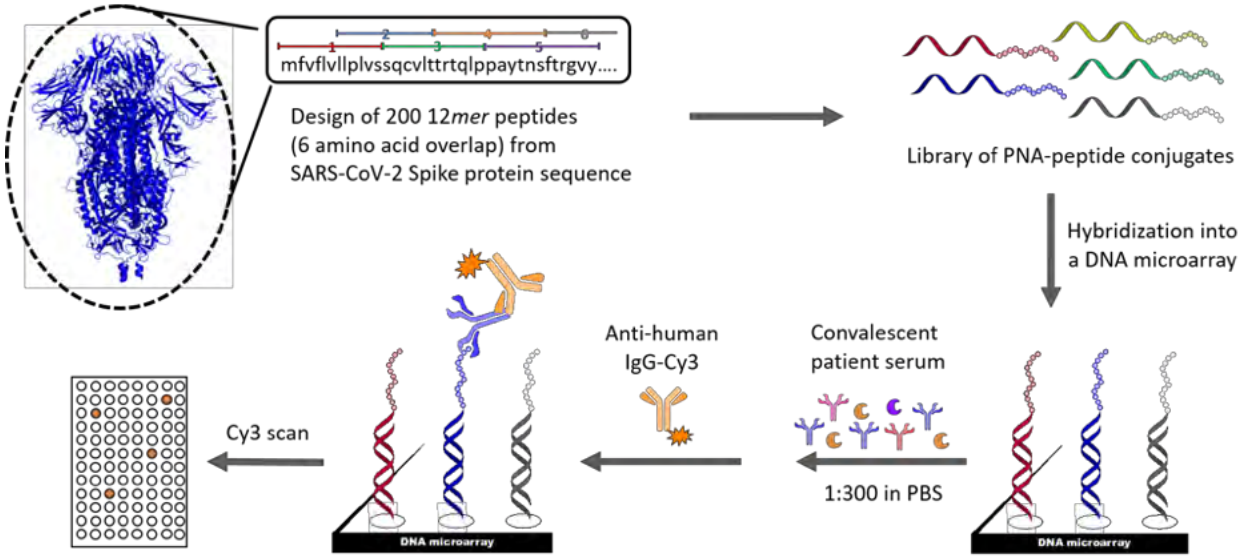
On December 2019 a novel infectious disease causing pneumonia was identified in the city of Wuhan (China). This new infectious disease (COVID-19) caused by severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) has significantly impacted the economy, education and life of many countries in the world. The World Health Organization declared it a pandemic on the 11th March 2020.



Mapping the epitope response of the immune system against the virus is of vital importance since it can lead to potential vaccines, more specific serological tests and to neutralizing antibodies which could be finally used as a treatment. In this work,[1] using a peptide microarray, we have identified 3 immunodominant regions on the spike protein which are only present in COVID-19 convalescent patients' sera which could lead to potential neutralizing antibodies.



Reference:

1. Farrera-Soler L., Daguer J-P., Barluenga S., et al. *PLoS ONE*. **2020**, *15(9)*, e0238089.

Access to optically active 7-membered rings by a 2-step catalytic sequence

Michele GARBO

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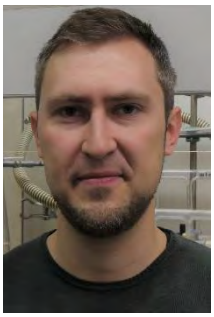
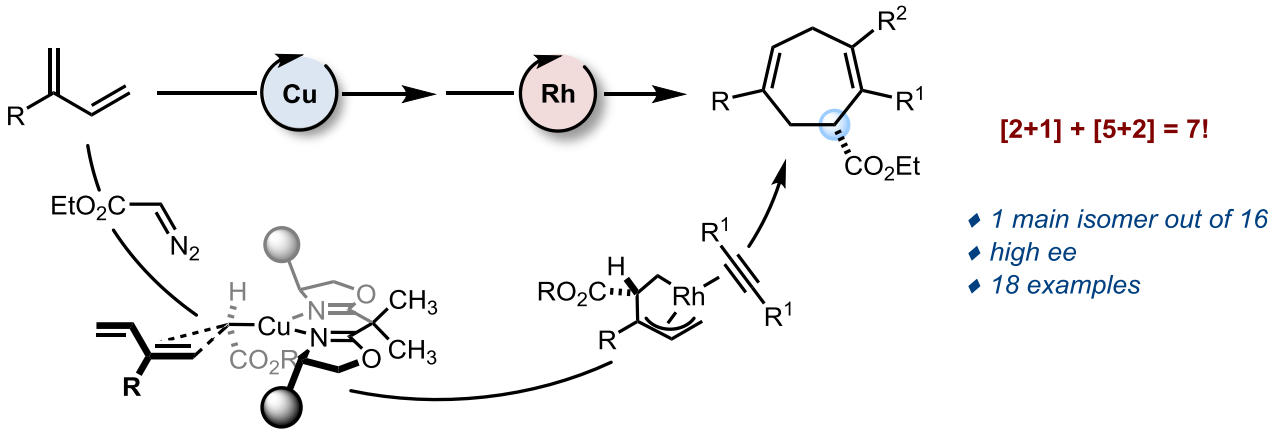


Seven-membered carbocycles are a common motif in natural products and biologically active c ompounds, in particular within the terpenoid family.[1] Methodologies for their stereoselective preparation are not nearly as much developed as for six-membered cycles and often rely on cycloaddition reactions. Despite fundamental advances over the last two decades, there is no report to date of an intermolecular and enantioselective [5+2] cycloaddition.[2,3] Its development would constitute an undoubtedly useful addition to the current portfolio and provide access to valuable stereochemically complex 7-membered rings. Herein, we report the successful realization of this objective.[4]



Our approach is based on a multimetallic cyclopropanation/cycloaddition catalytic sequence that provides access to optically active 7-membered rings starting from readily available conjugated 1,3-dienes. In the first step, the Cu-catalyzed cyclopropanation of branched dienes generates vinylcyclopropanes in high yield, regioselectivity and enantioselectivity albeit in modest *cis*/*trans* diastereoselectivity. The stereoconvergent nature of the subsequent Rh-catalyzed [5+2] cycloaddition with alkynes overrides this apparent limitation and affords preferentially one 7-membered ring out of the 8 or 16 possible stereoisomers that can be theoretically generated. The final regioselectivity and enantioselectivity are high in the majority of cases. The method is versatile and tolerates a broad range of functional groups.

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SH3 domains regulate spatio-temporal assembly of actin nucleation promoting factors in endocytosis

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In clathrin-mediated endocytosis, cells actively take up molecules from their surface by engulfing and finally pinching off a piece of their plasma membrane. In budding yeast, this process is mechanically dependent on actin polymerization. The actin filaments are anchored to adaptor proteins and lateral nucleation of new filaments leads to growth of a branched actin network and thereby to a directed force transmission. Most actin nucleation promoting factors (NPFs) have proline-rich motifs (PRMs) and/or so-called SH3 domains that can specifically identify and bind PRMs.

In this study, we used different live cell fluorescence microscopy approaches to find out to what regard SH3 domains mediate assembly of actin NPFs. SH3 domains of adaptor complex proteins recruit actin NPFs to endocytic sites. When actin polymerization starts, actin filament associated SH3 proteins down-regulate actin NPF binding to the adaptor complex. This ensures localization of actin NPFs to the actin nucleation control zone. Furthermore, we can show that the early SH3 domains are dominant actin NPF binding competitors against SH3 domains of plasma membrane sensing proteins. Interestingly, these SH3 domains bind to their interaction partners in the very late stage of endocytosis and our experiments indicate that they mediate disassembly of actin NPFs.

In summary, actin NPF regulation is temporally and spatially coordinated by SH3 domains. The SH3 interaction network is a very dynamic actin NPF regulation system, potentially promoting feedback-driven actin NPF control.

Ultra-sensitive pH measurements with a coulometric principle

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The pH glass electrode is measured in terms of its potential difference against a standardized reference electrode containing a 3 M KCl electrolyte in contact with the sample through a liquid junction. Unfortunately, some modern applications require sensitivities that cannot be achieved with potentiometric probes, which give reproducibilities on the order of mpH units. We have recently reported that pH measurements from potentiometric probes can be made dramatically more sensitive by operating in a capacitive detection mode. The unique feature of this work is the use of electronic capacitor component placed in series with a H^+-selective electrode (H^+-ISE), in a so-called "constant potential capacitive readout".[1] The potential of the cell is kept at a constant value by a potentiostat. A change of ion activity results in a potential change at the sample−membrane interface of the pH probe. Since the cell potential remains constant, the change is compensated by charging an electronic capacitor with the same potential of opposite sign until the new equilibrium state is established. Alternating of reference and sample solutions results in capacitive transient current response. In this way, pH changes of 0.001 pH units are easily quantified with a precision that is much better than that of potentiometric measurements. Integration of transient current gives a charge as readout signal that is proportional to the logarithm of hydrogen ion activity. Larger capacitance results in greater charge proportional to the equation of $Q = C \cdot s \cdot \Delta pH$, where C is the added capacitance and s is Nernstian slope, typically 59.2 mV. Very high reproducibilities of 28 µpH and 67 µpH were achieved for buffer and stabilized seawater samples, respectively.

Solid-contact ion-selective electrodes (SC-ISEs) were introduced aiming for improved practical use. Here, the inner filling solution of the electrode is replaced by an ion-to-electron transducing material, *e.g.* functionalized-single was carbon nanotubes (f-SWCNTs) and poly(3-octylthiophene) (POT). When using the capacitive characteristic of the transducing material alone, the coulometric readout gives long response times and drifting baseline current. This limitation is overcome by again placing an electronic capacitor in series to the SC-ISEs to dominate the overall cell capacitance.[2] Kirchhoff's law is applied to estimate the total capacitance for two capacitors placed in series. The response time for the transient current to get back to the equilibrium state was now dramatically reduced (to less than 10 s) and the current drift was mostly eliminated. The range of added capacitance compatible for SC-ISEs is up to 100 µF for f-SWCNTs, while POT layers require a narrower range of 1−4.7 µF.

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Separating boundary potential changes at thin solid contact ion transfer voltammetric membrane electrodes

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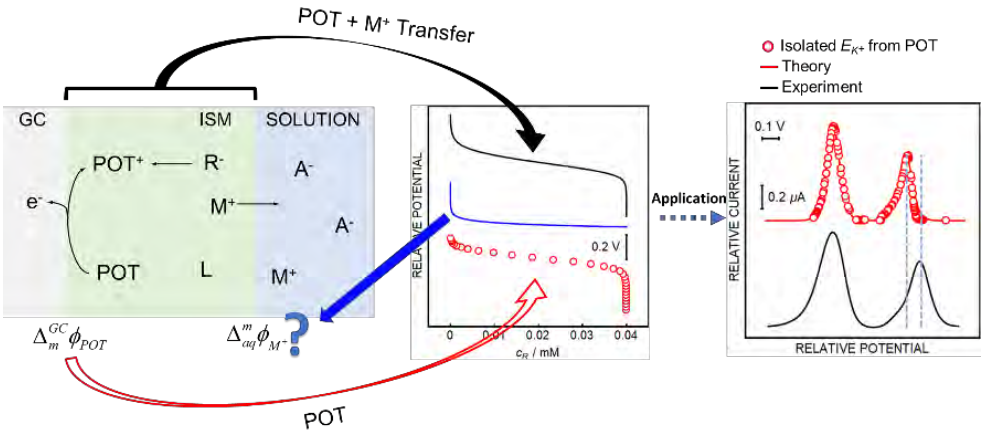
Ion transfer at composite ion-selective membrane is difficult to study because the traditional experimental conditions involve thermodynamic and kinetic processes together. A thin ion-selective membrane system provides us with an accessible way to avoid kinetic limitation and we can investigate ion transfer voltammetry on solid contact membrane electrode by approaching an ideal situation.

Here we dope a redox molecule, lipophilic (1-dodecyl-1H-1,2,3-triazol-4-yl)ferrocene,[1] in a thin membrane containing a cation-exchanger to study the separate inner and outer boundary potential changes using a monovalent reference cation for ion transfer. Based on the electroneutrality, the cation is transferred out of the membrane when the redox molecule is electrochemically oxidized from the electrically neutral form. It follows the equilibrium conditions as the hypothesis and the results agree well with thermodynamic theory.[2]

For a given membrane composition, the experimental full peak width at half maximum is nearly the same as the theoretical value, 0.110 V, and the observed linearity between peak current and scan rate indicates that this model system does not involve diffusion limitations. The charge converted from the integration of ion transfer current is then correlated to the available ion-exchanger quantity, which is plotted as a function of potential. Then, with the calculated potential change from the reference ion, the potential change for the ion-to-electron transducer can be isolated from the experimental data. As a practical example, the common transducing layer poly(3-octylthiophene) (POT) was characterized in this manner. It shows a non-linear relationship of potential as a function of charge. The approach allows one to isolate the ion transfer wave for a range of solid-contact ion-selective membranes.



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Oligomers of cyclic oligochalcogenides for enhanced cellular uptake

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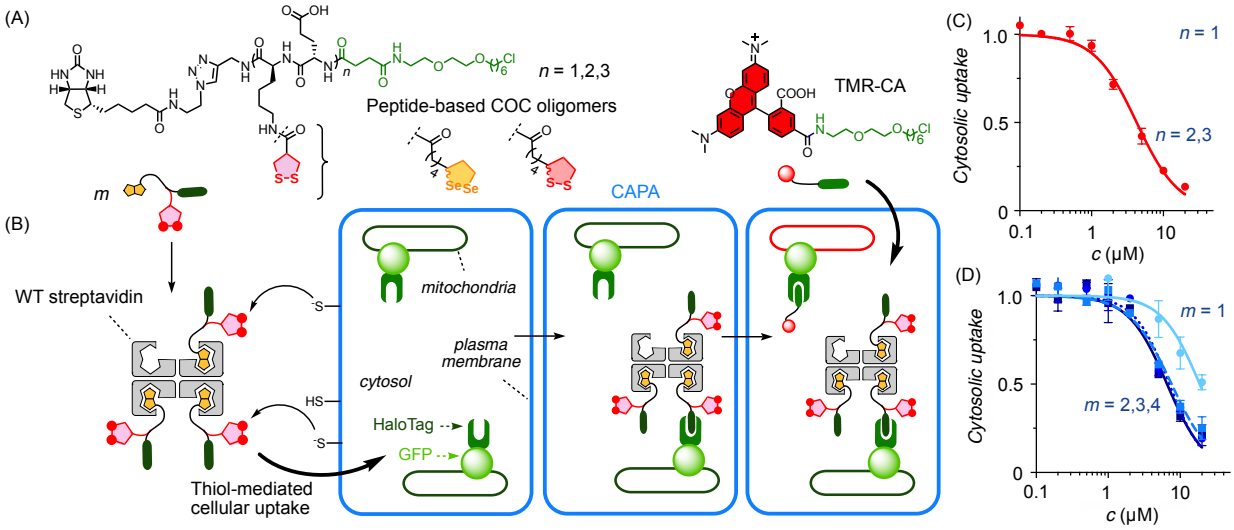
Recent development of polymeric linear disulfides, known as cell-penetrating polydisulfides (CPDs),[1] and monomeric cyclic oligochalcogenides (COCs)[2] as powerful thiol-mediated uptake transporters called for the introduction of COC oligomers.[3] To explore COC synergism and possible oligomer effects emerging from multivalent interactions with membrane thiols (Figure 1B), we designed a peptide-based transporter, with *n* alternating units (*n* = 1, 2 or 3) containing a lysine and a glutamic acid, to attach the COCs and to increase solubility, respectively (Figure 1A). Asparagusic, lipoic and diselenolipoic acids were chosen to examine potential effects of the COCs themselves. Next, the N-terminus was capped with a chloroalkane, to report on cytosolic uptake using the invaluable chloroalkane penetration assay[4] (CAPA, Figure 1B). In contrast, supramolecular oligomeric effects could then be assessed by binding multivalent peptides to the tetrameric streptavidin using a biotin attached on the C-terminus of peptides by click chemistry. Consequently, *m* biotinylated peptides (containing *n* COCs) were bound to streptavidin (Figure 1B) and their uptake was systematically evaluated by CAPA. In general, efficiency increased from covalent monomers to dimers but saturated at trimer level (Figure 1C). On the supramolecular level, saturation occurred at dimer level (Figure 1D), displaying dimers of dimers as new lead structures for thiol-mediated cellular uptake. Latest results also proved a very remarkable sequence dependence of the peptide structure on cellular uptake.



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Single cell analysis unravels a perturbation of the cell cycle of the host *Dictyostelium discoideum* during infection by *Mycobacterium marinum*

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Mycobacterium marinum and *M. tuberculosis* are close relatives and share similar virulence mechanisms. They are able to manipulate the macrophage phagosome maturation pathway in order to establish a permissive compartment to proliferate. On the other hand, *Dictyostelium discoideum* is a social amoeba and a professional phagocyte with highly conserved cell-autonomous defence systems. It is a powerful model organism to study infection and dissemination of pathogenic mycobacteria.

The study of intrinsically heterogeneous populations can mask cell individuality and important phenotypic heterogeneities, potentially having causal impacts on the course of an infection. To correct this bias, the "InfectChip", a microfluidic device recently developed to perform long-term single-cell imaging of amoeba-bacteria interactions is used. This device enables the monitoring of the integrated history of the infection course and precisely map and quantitate the fates of the host and the pathogen during infection.

Previous work in the lab demonstrated that TOR, a key protein kinase involved in nutrient sensing and central control of cell growth and aging, is modulated during infection. In addition, RNAseq analysis revealed a downregulation of DNA replication, cell division and ribosome biogenesis. This cumulated evidence leads to the hypothesis that infection might affect the cell cycles of the host *D. discoideum* and the intracellular pathogen *M. marinum*.

Our result show that, in control conditions, the *D. discoideum* interdivision time in the InfectChip is around 8 hours, but the average interdivision time of infected cells is about 17 hours. Surprisingly, non-infected cells, which are either previously infected cells or potentially influenced by contact with bacteria also present a higher interdivision time (13 hours). In addition, preliminary analyses indicate that during host cell division, there is no evidence for a bias in the inheritance of the cytoplasmic bacteria. Interestingly, the effect of infection on the interdivision time appears to be damage-dependent. Indeed, cells infected with *M. marinum* with a non-functional ESX-1 secretion system (ΔRD1) have an interdivision time of 15 hours and even infection with dead mycobacteria leads to an interdivision time of 13 hours. Measuring the size of *D. discoideum* cells during infection **revealed** both a significant increase in the heterogeneity of growth rates and in in the probability of reaching a growth plateau before cytokinesis. Monitoring of the nucleus size and Histone H2B content reveals that the mechanism that retards cell division is likely linked to a nuclear division checkpoint.

Polydiacetylene-peptide interaction mechanism in mixed lipid systems

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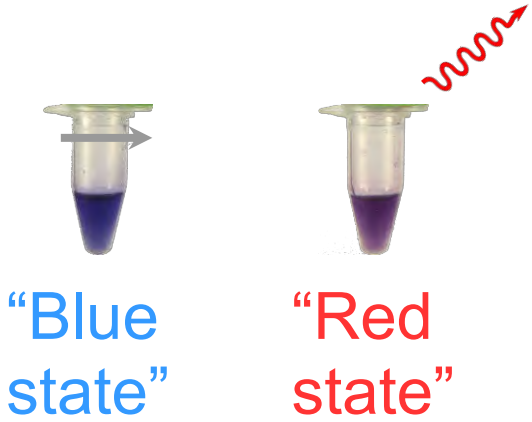
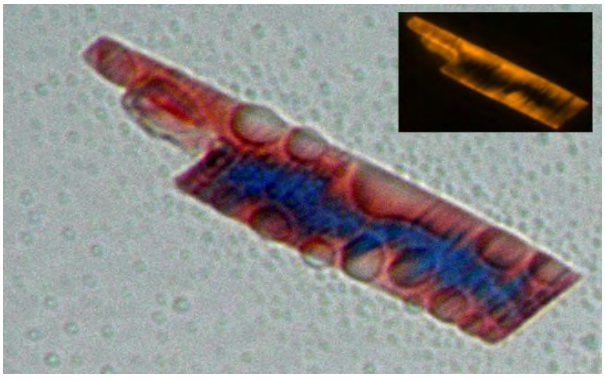
Opening mechanosensitive ion channels, cellular virus infection and killing bacteria by antimicrobial peptides, all involve an application of forces to the cell membranes. Characterization techniques such as giant unilamellar vesicle aspiration and force spectroscopy are able to extract information about the surface tension and binding forces respectively. However, until now there is no possibility to measure the local molecular forces in lipid bilayer. Therefore, the goal of this project is to develop a calibrated fluorescence probe for mapping forces in cell membranes by the mechanosensitive polymer polydiacetylene (PDA).

PDA is a popular mechanosensitive polymer, used as chromic and fluorescence biosensors for the detection of ions, ligands, bacteria and peptides. The current center of debate is the molecular mechanism of the PDA activation by these ligands, where how these biomolecules alter the PDA structure and thus change its optical properties are left unexplored. In this work, to clarify the mechanism of the PDA activation by peptides, we investigated the interaction between PDA and an antimicrobial peptide from bee venom, melittin, by fluorescence and atomic force microscopy. These microscopy techniques provide spatio-temporal resolution in contrast to the traditional spectroscopy technique used in previous works, which revealed unique interaction kinetics between the peptides and PDA. Our new approach lets us furthermore observe the phase transition of the PDA domains induced by peptides and compare it to classic differential scanning calorimetry technique.[1] Understanding the peptide-PDA interaction mechanism is the first step to engineer this material for the use as a peptide force sensor.





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Modular synthesis of mono and dicationic [4]helicenes with complementary biaryl atropisomerism

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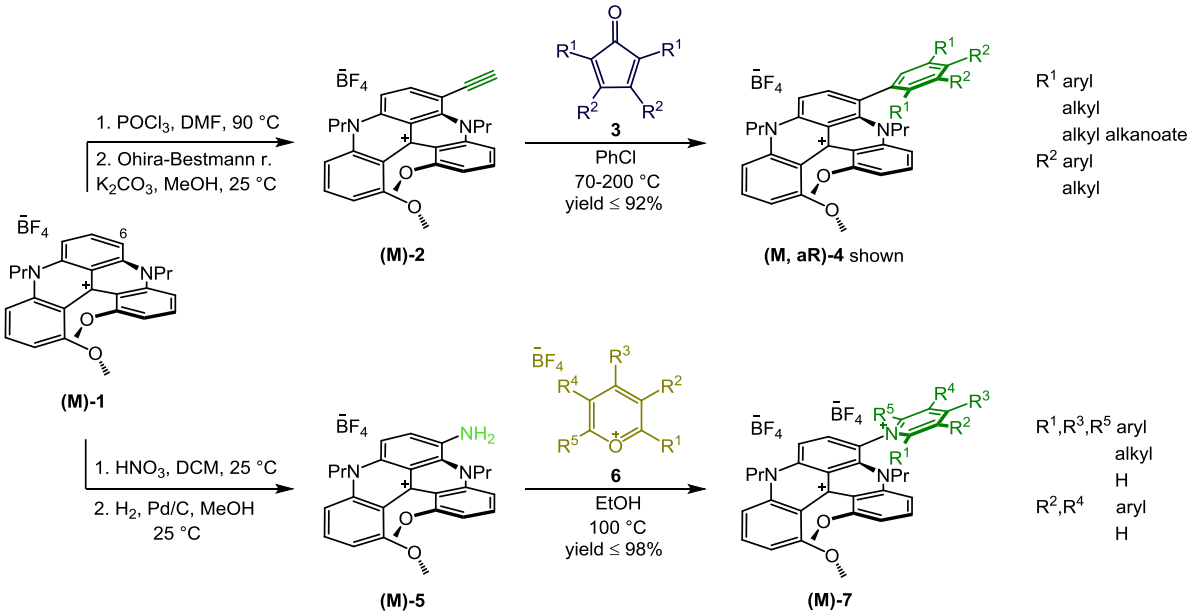
Helicenes are *ortho*-fused polycyclic aromatic derivatives that find many applications in asymmetric catalysis, molecular recognition and as molecular machines or chiral chromophores.[1] In this context, our group studies highly stable aza helicenium salts, which are inert in air and in strongly alkaline aqueous solutions despite their cationic nature.[2] In this work, diaza [4]helicene motif **1** was used as a core structure.[3] It possesses a helical chirality that was used to promote effective complementary biaryl atropisomerism.

In addition, **1** can be selectively functionalized with electrophiles at position 6.[4] Ethynyl derivative **2** can thus be made and used in sequences of Diels-Alder/retro-Diels-Alder reactions with cyclopentadienones **3**.[5] Derivatives of type **4** were synthetized by this approach with yields up to 92%. Some of compounds **4** are obtained with very high diastereoselectivity around the new biaryl bond (dr up to 25:1, ^{1}H NMR).



Furthermore, amino **5** was used in Katritzky reactions[6] with pyrylium salts **6** forming dicationic pyridiniums **7** (≤ 98%, dr ≤ 28:1). For both sets of compounds, optical and chiroptical properties were investigated.

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Total synthesis of goyazensolide and identification of the first importin-5 inhibitor

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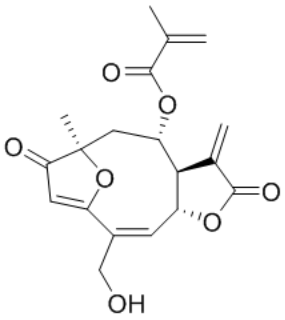
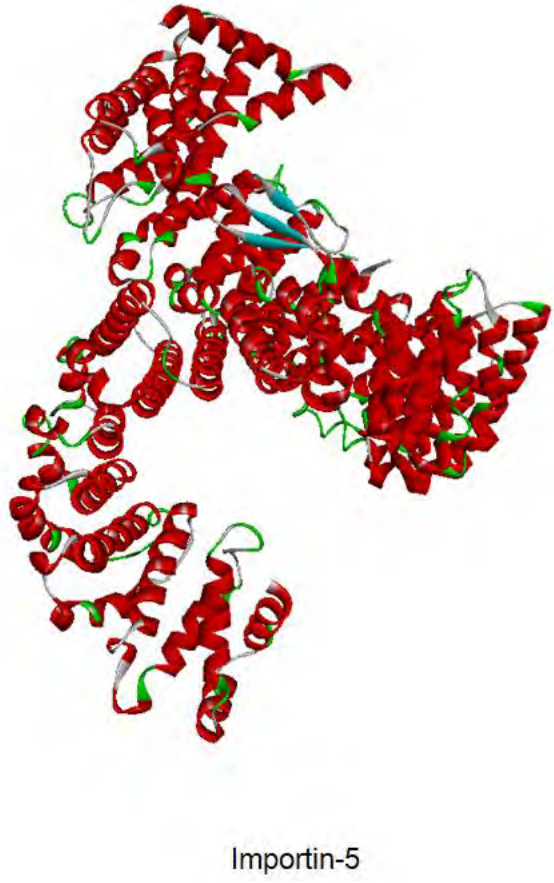
Sesquiterpenes are a rich source of covalent inhibitors with a long history in traditional medicine and include several important therapeutics and tool compounds. Herein we report the total synthesis of goyazensolide via a build/couple/pair strategy. Using an alkyne-tagged cellular probe and proteomics analysis, we discovered that goyazensolide selectively targets the oncoprotein importin-5 (IPO5) for covalent engagement. We further demonstrate that goyazensolide inhibits the translocation of RASAL-2, a cargo of IPO5, into the nucleus and perturbs the binding between IPO5 and two specific viral nuclear localization sequences.



Goyazensolide



Importin-5

Peptide stapling with Anion-π Catalysts

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Cation-π interactions were reported to play a central role in variety of biological processes. One of the most important examples is the contribution of cation-π interactions in the enzymatic biosynthesis of terpenes and steroids using polyenecyclases.[1] In contrast, the complementary, counter interactions – anion-π interactions rarely occurs in nature. Even though, anion-π catalysis has emerged recently as a promising tool for performing a wide range of chemical transformations.[2] Most examples are limited only in the context of organic synthesis. To examine the potential applications of anion-π interactions in more biologically relevant processes, we incorporated anion-π catalysts into several short peptides and evaluated their activities in catalysis.[3]

The matching in dimension of aromatic electron-deficient naphthalenediimides (NDIs) with one turn of an α helix gives the possibility of using NDIs not only as a platform for catalysis but also as a linker for stabilizing peptide secondary structures. Introduction of tertiary amine bases in amino-acid sidechains initiates anionic transition state above π-acidic NDI surface (Figure 1). The addition of malonic acid half thioesters to enolate acceptors (MAHT reaction, Figure 2) was chosen to evaluate synthesized catalysts. According to this benchmark reaction, anion-π catalysis next to peptides occurs with record chemoselectivity but weak enantioselectivity. The importance of peptide secondary structure in catalysts' design was proven by the decreasing of catalytic activity when mutations were introduced into peptide structures (changing position of the amine base, using homocysteine bridges, fastened and loosened α-helix turns, removing NDI linker). Elongation of peptide turn into short α helices significantly increases activity in accordance with the increasing of structure organization. These results encourage integration of the NDI-stapling motif into larger protein structures for different purposes, including anion-π enzymes.

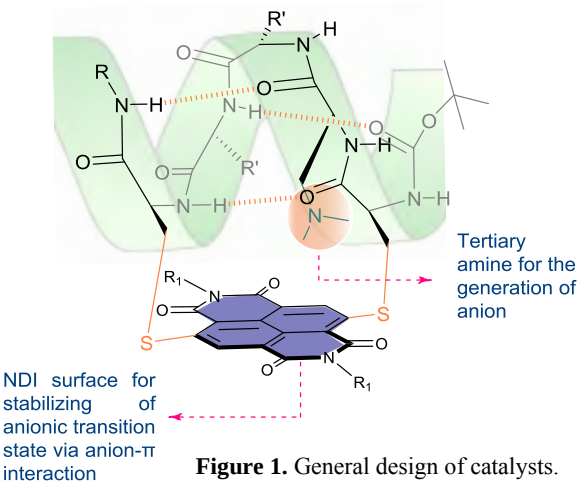

Figure 1. General design of catalysts.

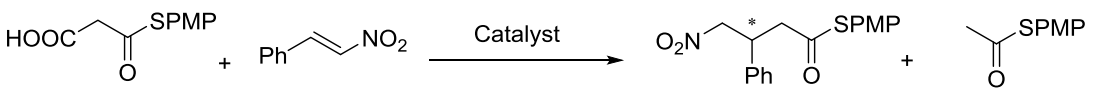

Figure 2. MAHT reaction.

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Frozen Density Embedding Theory: What densities can one use? What errors can one expect?

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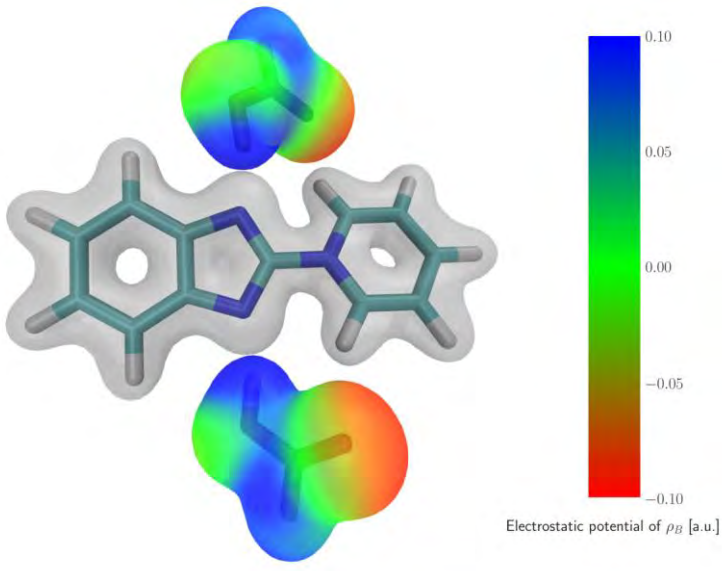
In chemistry and physics, modelling of a molecule in a specific environment, be it a solvent, a crystal, or a protein, is often crucial. The unfavourable scaling of quantum chemical methods makes this a difficult task. Multi-scale methods tackle this by using different levels of theory in different regions of the space.

Among them, Frozen Density Embedding Theory (FDET)[1-3] is an exact theory based on DFT where the environment is modelled via its electron density and which allows a quantum-level treatment of the whole system.

The sources of error in FDET-based calculations are known, but the extent of their effect is not easy to predict. Furthermore, the approximations applied in the equations do not have a one-to-one correspondence to chemical phenomena.

One of the advantages of FDET is that several choices are possible for the density used to model the environment, each with its own advantages, disadvantages, and range of applicability.

An overview of the possible choices for the environment density[3-6] will be given, and their link to the sources of errors in the equations and the resulting error in calculations will be discussed.



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Amplification of enantiomeric excess by dynamic inversion of enantiomers in deracemization of Au_{38} clusters

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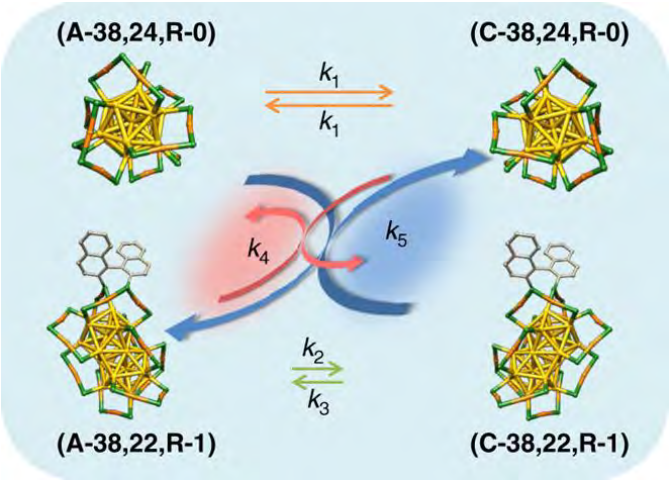
The noble metal nanocluster as kind of ultrasmall materials and bridge the gap between nanoparticle and molecular, has attracted interesting during the past decades. Among the subsets, gold nanoclusters because of the scheduled synthesis, high stability and special properties, have been employed into wide applications such as catalyst, as sensor and for medical therapy.[1-3]

$Au_{38}(2\text{-PET})_{24}$ (2-PET = 2-phenylethylthiolate) is an initial chiral nanocluster, and also important model for investigating chiral properties of gold clusters. The first enantioseparation of this cluster was succeeded by chiral HPL chromatograph.[4]

Recently, we found that the balance between left- (anticlockwise) and right- (clockwise) handed enantiomers of $Au_{38}(2\text{-PET})_{24}$ can be broken by adsorbing a small amount of a chiral molecule in its ligand shell. We studied the amplification of enantiomeric excess of the $Au_{38}(2\text{-PET})_{24}$ cluster, which may also called deracemiazation.[5] At 70 °C, the system evolves towards the anticlockwise clusters at the expense of the clockwise antipode. It is shown that the interplay between the diastereospecific ligand exchange, which introduces selectivity but does not change the A/C ratio, and the fast racemization of the $Au_{38}(2\text{-PET})_{24}$ is at the origin of this observation.[6]



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THE SCHOOL OF CHEMISTRY AND BIOCHEMISTRY

The *Section de chimie et biochimie*, University of Geneva, offers a top tier training environment that results in a highly competitive expertise. An increasing number of diplomas (now *ca.* 70 per year) at the Bachelor, Master and Doctoral levels in chemistry, biochemistry, and chemical biology, are being delivered to foreign and Swiss students.

The *Section de chimie et biochimie* produces about 200 publications per year and plays host to the National Centre of Competence in Research *Chemical Biology*. The research themes encompass most essential areas of fundamental molecular and biomolecular sciences:

- Elucidation and modelling of the behaviour of complex molecules on ever-shorter time scales.
- Elaboration of new supramolecular architectures on the nanometre scale with promising microscopic and macroscopic properties.
- Development of analytical techniques surpassing today's frontiers of precision, in order to identify ultra-trace species in complex environments.
- Development and optimisation of alternatives to fossil fuels as sources of energy.
- Development of highly selective and environmentally benign methods of organic synthesis.
- Work towards an understanding of the biomacromolecules involved in the processes governing the living world at the interfaces between chemistry, biology and medicine.

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NCCR CHEMICAL BIOLOGY

Swiss National Centre of Competence in Research Chemical Biology
Hosted by the School of chemistry and biochemistry
http://nccr-chembio.ch

Chimiscope UNIGE

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SCS
Swiss Chemical Society

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LS2
Life Sciences Switzerland

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