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## The 3rd Transatlantic Frontiers in Chemistry Symposium

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The 3rd Transatlantic Frontiers of Chemistry Symposium, held at Kloster Seeon in Germany on August 8–11, 2013 brought together 60 early-career chemistry academics from Germany (20), UK (18), USA (16) and for the first time, Brazil (6). The aims of the conference were to enable international, multidisciplinary networking and collaboration opportunities in the relaxing settings of a beautiful old monastery by a lake (Figure 1). The program included a diverse

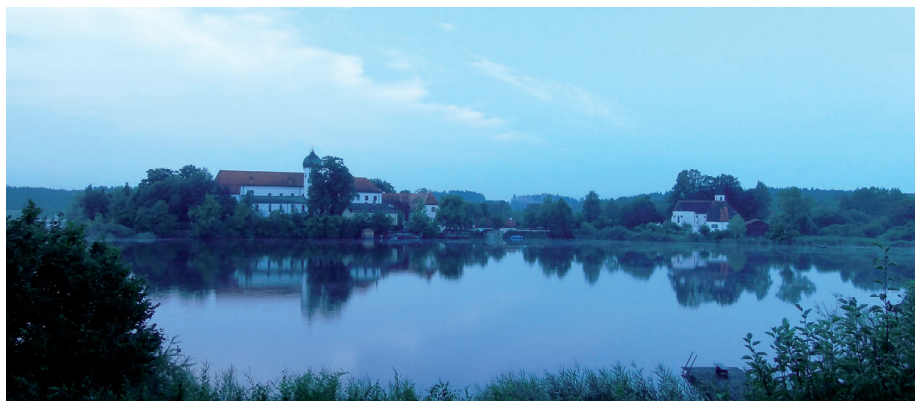


Figure 1. Kloster Seeon: the setting for the 3rd Transatlantic Frontiers of Chemistry Symposium; photo: A. Alexandrova.

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set of topics that were grouped into related sessions. The meeting started with an address by representatives of the organizing committee (Gesellschaft Deutscher Chemiker, American Chemical Society, and Royal Society of Chemistry; Figure 2), followed by remarks from the conference chair, Prof. Dr. Hans-Dieter Arndt: “you are in one of the most beautiful parts of Germany, with plenty of things to do and see in the area. Unfortunately you will be spending quite a bit of your time during the coming days in this lecture theater...” (Figure 3). Each delegate was then asked to stand up and introduce themselves, and it was immediately clear, that despite the involvement of only four national chemical societies, this was a truly international conference, with multiple countries represented; a running theme of this section was that most delegates were dual nationals and many were working in a different country from which

they had done their first degree. The international composition of the meeting was further commented upon by Prof. Dr. Helmut Schwarz (President of the Humboldt Foundation) during his after dinner lecture, as being in keeping with Alexander von Humboldt’s vision of science without borders (Figure 4).

### Functional Materials and Catalysts

The scientific program opened with the first session on functional materials, which promise solutions to contemporary energy conversion and storage problems through efficient catalysis, CO<sub>2</sub> sequestration, and more. Through the most advanced methods of material synthesis, design, and



Figure 2. Some of the invited guests and members of the organizing committee for the symposium: Back row left to right: Hans-Georg Weinig (GDCh), Christian P. R. Hackenberger (Berlin), Eckhard Ottow (Vice President, GDCh), Helge B. Bode (Frankfurt), Daniel B. Werz (Braunschweig), Peter Gölitz (Wiley-VCH), Sven Schneider (Göttingen), Helmut Schwarz (President of the Alexander von Humboldt Foundation), Hans-Dieter Arndt (Jena), Robert Parker (CEO, RSC), Bradley D. Miller (ACS), James Hutchinson (RSC); front row left to right: Marinda Li Wu (President, ACS), Eva Wille (Wiley-VCH), Kelen Fureigh (ACS), Rachel O'Reilly (Warwick), Danica Fujimori (San Francisco), Timo Jacob (Ulm), Anastassia N. Alexandrova (Los Angeles), Edward Anderson (Oxford); photo: A. Wirt and H.-G. Weinig (GDCh).

characterization, participants targeted the key applications relevant to modern problems such as photovoltaics and solar energy conversion, heterogeneous catalysis, electroanalytical chemistry, and even drug delivery.

**M**esoporous materials are currently undergoing a boom, and received deserved attention at the TFOC Symposium. Mircea Dincă (Massachusetts Institute of Technology) detailed his vision to make metal-organic frameworks (MOFs) electrically conductive. Such MOFs would be an exciting type of new material, since they would be thermally insulating (by definition, since they are mesoporous), and at the same time electrically conductive. By intertwining MOFs with molecular conductors, charge mobility surpassing that of many organic semiconductors was achieved. Departing from MOFs, Bettina Lotsch from the Max Planck Institute



Figure 3. The conference chairman, Hans-Dieter Arndt, Friedrich-Schiller University Jena; photo: A. Wirt and H.-G. Weinig (GDCh).

in Munich has designed purely covalent organic frameworks (COFs), which are exceptionally light materials accessible through conventional organic synthesis routes. Mesoporous material architecture was further masterfully explored by Delia Milliron (Lawrence Berkeley National Laboratory). Her group develops methods for arranging colloidal nanocrystals into electrochemically active composites. Strategic preparation of such materials allows for the appearance of interesting properties, such as ionic conductivity and dynamical optical response, given by both the overall material architecture and the properties of its building blocks.

**O**n the solar cell front, Thomas Hamann (Michigan State University) focused on reducing the cost of materials in use. His strategy is the design of efficient dye-sensitized solar cells based on nanoparticle  $\text{TiO}_2$  electrodes. Matt Law (University of California, Irvine) aims to utilize colloidal films of quantum dots to do the same job. The key aspect here is to make these systems conductive; electrical conductivity is achieved via inter-QD tunneling, which can be manipulated through particle size, doping, and inter-particle spacing.

**R**achel O'Reilly (Warwick University) has found a way to attach small fluorescent labels, dithiomaleimides, to polymers, which allowed for tracking polymer conformations in a variety of applications, such as drug delivery and formation of nanostructures. Even DNA can be labeled in this way without the disruption of its structure, which provides an amazing insight into its structure and function in a variety of contexts.

**O**verall, the materials group provided a set of "appetizers" from their diverse and rapidly expanding field, teasing the TFOC participants and guests with what materials can do, and how scientists can strategically design them for desired properties.

**T**he next session on metal-centered catalysis included three fundamental studies on transition-metal complexes with nitrogen- or carbon-based ligands. In the context of using ammonia as an energy carrier, Sven Schneider (University of Göttingen) presented work on square-planar iridido nitrido complexes, their electronic characterization and the coupling of such species to form dinitrogen. Studying close chemical relatives, namely highly reactive copper tosyl nitrene complexes, Kallol Ray (Humboldt University Berlin) showed



Figure 4. President of the Humboldt Foundation Helmut Schwarz who gave the after-dinner lecture entitled “Overcoming Frontiers in Science: Messages and Recommendations of the Humboldt Foundation”; photo: A. Wirt and H.-G. Weinig (GDCh).

that these intermediates can be trapped by Lewis acids such as scandium triflate, allowing the complete characterization of such species at  $-90^{\circ}\text{C}$  and subsequent studies on their chemical reactivity. Viktoria Gessner (University of Würzburg) rounded out this area of research with her studies on geminal-dianion-derived carbene ligands as electronic alternatives to the well-known Fischer- and Schrock-type carbenes. The application of transition-metal complexes to organic synthesis was the main focus of talks by Igor Larrosa (Queen Mary University of London) and Stephen Thomas (University of Edinburgh). Larrosa showed how a combined C–H activation/decarboxylation strategy can be employed for formal *meta*-selective direct arylation reactions. In some contrast to these decarboxylation reactions, Thomas described the synthesis of  $\alpha$ -aryl carboxylic acids from styrene derivatives, using bench-stable  $\text{Fe}^{\text{II}}$  precatalysts,  $\text{CO}_2$ , and  $\text{EtMgBr}$  as hydride source. The focus of the three remaining speakers was on nano-sized structures. Pedro Camargo (University of São Paulo), one of the six participants from Brazil, presented solution-phase approaches to produce metallic nanoparticles with controlled shapes, which find application as surface-enhanced Raman scattering substrates or catalysts. Yu Huang (University of California, Los Angeles) then focused on molecular details of inorganic–organic interface interactions. She uses facet-specific peptide sequences to produce Pt nanocrystals with predictable shape and analyzes the binding mechanism by experimental and theoretical studies. Finally, Erwin Reisner (University of Cambridge) showed how enzymes and synthetic catalysts can be integrated in nanostructured materials. More specifically, he described the immobilization of hydrogenases or cobalox-

imes on Ru-dye-sensitized  $\text{TiO}_2$  and the use of these hybrid materials for photocatalytic  $\text{H}_2$  production.

### Chemistry *In Silico*

A tight theory and computation group presented two vibrant sessions at the TFOC Symposium. Because of the multidisciplinary nature of the meeting, the theorists made a significant effort to come down to earth and explain to non-experts what they do and how they do it. Perhaps for this reason, the “*out of silico*” participants were able to greatly enjoy the *in silico* part of the meeting. Chemistry *in silico* exists in a wide spectrum between isolated formal theory and pure computation done as a “cherry on top of an experimental cake”, with the healthy middle being the most exciting, as exemplified by the speakers. Theory not only provides explanation for existing experimental observations, but also gives new chemical and physical insights inaccessible through the experiment, and also pushes the development of new enabling methodologies. The presentations clustered around several modern challenges for theory: non-equilibrium and quantum effects in chemical reactions (David Glowacki, University of Bristol; Johannes Kästner, University of Stuttgart; Thomas Miller, California Institute of Technology), proper many-body description of weak interactions (Alexandre Tkatchenko, Fritz-Haber-Institut der Max Planck-Gesellschaft; Lasse Jensen, Pennsylvania State University), and multi-scale dynamic treatment of complex systems (Anastassia Alexandrova, Thomas Miller, California Institute of Technology, Timo Jacob, Ulm University); as well as key challenging applications: reaction mechanisms and dynamics (Robert Paton, University of Oxford; Johannes Kästner, University of Stuttgart; David Glowacki, University of Bristol; Anastassia Alexandrova, University of California Los Angeles, UCLA; Thomas Miller, California Institute of Technology, Timo Jacob, Ulm University), electron and proton transfer in biology (Thomas Miller, California Institute of Technology), enzymology and enzyme design (Anastassia Alexandrova, UCLA; Thomas Miller, California Institute of Technology, Robert Paton, University of Oxford), protein aggregation (Birgit Strodel, Research Centre Jülich), electrochemistry (Timo Jacob, Ulm University), and molecular surface-enhanced Raman spectroscopy (Lasse Jensen, Pennsylvania State University).

Presentations stirred the audience away from the conventional wisdom of textbooks. For example, the notion of classical paths in reaction mechanisms and dynamics was challenged and broken, twice. First, Johannes Kästner from the University of Stuttgart opened the show with a demonstration of how the inclusion of quantum-mechanical tunneling in identifying reaction mechanisms could be crucial, and without it the wrong preferred mechanism could be predicted. David Glowacki from the University of Bristol brought further surprises: dynamics of chemical reactions can be such that the full vibrational relaxation of every reaction in-



intermediate is often not achievable before the system crosses the next barrier on the reaction path. This again challenged the classical perspective of reaction mechanisms, in this instance from a non-equilibrium statistical mechanical standpoint.

**I**n silico research also yielded new controversial chemistry. Anastassia Alexandrova (University of California, Los Angeles) challenged the catalytic activity of natural enzymes. The natural evolution of enzymes simultaneously optimizes many variables besides catalysis (protein stability in living cells, competition of different metals for metal binding sites, need for channels and chaperones, and the utilization of bio-available and nontoxic components), whereas *in vitro*, most of these constraints are removed, and therefore, catalysis should be improvable. In this spirit, Alexandrova challenged the choices Nature has made for metals in certain biological enzymes, and demonstrated and explained some surprising effects on their functionality and activity of metal replacement. Thomas Miller, California Institute of Technology addressed the standing question of how and if quantum processes, such as electron or proton transfer in proteins, couple to protein dynamics. The answer, he suggested, is due to differences in time scales, they barely do. Instead, the protein undergoes its native dynamics until it reaches a configuration favoring the quantum process, and then the latter happens rapidly with a minimal coupling to the surroundings.

**O**n the methodological front, the need for a multi-scale treatment of complex chemical systems was emphasized. Techniques ranging from quantum electrodynamics, to path-integral molecular dynamics (MD), *ab initio* electronic structure calculations, classical MD, coarse graining, and various application-based marriages of all of the above were developed and virtuously used by the young researchers. Timo Jacob from Ulm University further advocated for the reactive force field MD, as a predictive tool for the mechanisms of electrochemical reactions in particular, which can be complex and proceed through multiple possible pathways. Additionally, Alexandre Tkatchenko (Fritz-Haber-Institut der Max Planck-Gesellschaft) warned us against the traditionally used pair-wise potentials for the description of van der Waals interactions, and offered a comprehensive and affordable solution for the inclusion of many-body effects.

**A**s a finale to the *in silico* session, David Glowacki introduced his Danceroom Spectroscopy initiative. A classical MD simulation of molecules is run and visualized on a large screen. Real people (in the museum or a stadium) can then move and dance near the exhibit, and their motion is fed back into the MD simulation as an additional term in the Hamiltonian. Thus people stir the molecules in real time and observe the result on the screen, in what is the World's first large-scale interactive MD experience. Glowacki's exhibition is now touring the world, and was featured at the

London Olympics in 2010. At the TFOC Symposium, dance-room spectroscopy generated an excitement and a rain of jokes, such as a proposal to use the approach to find out how much hotter heavy metal concerts are as compared to other kinds of people gatherings.

## Synthesis and Biosynthesis

**I**n the session on synthetic methods development, Diogo Lüdtke (Federal University of Rio Grande do Sul, Porto Alegre) presented his work on enantioselective arylation reactions of aldehydes, which are based on mixed aryl alkyl zinc compounds as arylating agents and chiral amino alcohols as ligands. High diastereoselectivities could be achieved in the reaction of achiral aryl alkyl zinc reagents with chiral substrates that were derived from carbohydrates or amino acids. Fascinating twists on cycloisomerization and -addition reactions were then reported by Christophe Aïssa (University of Liverpool), Edward Anderson (University of Oxford), and Tehshik Yoon (University of Wisconsin-Madison). In the transition-metal catalyzed carbon-carbon bond activation reactions discussed by Christophe Aïssa, intramolecular hydroacylation of asymmetrically substituted alkylidenecyclobutenes lead to cyclooctenone derivatives. The mechanism of these Rh<sup>I</sup>-catalyzed rearrangement reactions was studied by isotope-labelling experiments, and up to three carbon-carbon bond cleavage steps were found. Transition-metal-catalyzed cycloisomerizations are also the focus of the work by Edward Anderson, whose interests center on ynamides and enynamides. Palladium-catalyzed cyclizations of enynamides (prepared from *N*-tosylated ynamides) lead to the synthesis of various nitrogen heterocycles with high stereoselectivity. Tehshik Yoon, on the other hand, performs diverse cycloaddition reactions using visible light photocatalysis, thus rendering the use of ultraviolet light, which is commonly employed in photochemical syntheses, unnecessary. He presented his work on diastereoselective [2+2] enone cycloaddition reactions using transition-metal photocatalysts such as Ru(bipy)<sub>3</sub> in combination with a Lewis acid. The final two speakers studied substitution reactions of allylic compounds. Ai-Lan Lee (Heriot-Watt University Edinburgh) reported the direct allylic etherification of allylic alcohols with likewise unactivated alcohol nucleophiles. These Au<sup>I</sup>-catalyzed reactions proceed via an S<sub>N</sub>2' mechanism and feature high stereoselectivity (*E*) as well as broad substrate scope. The final speaker (Nuno Maulide, MPI Mülheim) had already impressed the audience on the previous day with this formidable mastery of the piano. Accompanied in part by an opera singer (the wife of Timo Jacob), Maulide presented works of various composers including Bach and Händel and introduced these pieces with insightful comments on the respective composition techniques. In his *scientific* contribution, he then demonstrated his equal mastery of allylic substitution reactions on cyclobutene derivatives, including deracemization and de-epimerization strategies.

The synthesis and biosynthesis session featured basic research studies by Niklaas Buurma (Cardiff University), Stefan Huber (Technische Universität Munich), and Steven Goldup (Queen Mary University of London). Buurma described the development of complex data analysis methods to dissect multiple-coupled equilibria, for example, in isothermal calorimetric titrations. The data analysis approach was illustrated by the binding of  $\pi$ -conjugated heteroaromatics like terthiophenes to DNA. Calorimetric titrations were also employed by Huber to study the binding behaviour of halogen-based Lewis acids to halides. These results enabled the use of such halogen-bond donors as organocatalysts in a carbon-carbon bond-forming reaction. Goldup presented his approach to construct mechanically planar chiral rotaxanes by a chiral auxiliary approach; the absolute stereochemistry of the rotaxane, which comprises an achiral macrocycle and an achiral thread, was unambiguously assigned. The use of complex rearrangements in organic synthesis was then illustrated by Daniel Werz (University of Braunschweig) and Tanja Gaich (University of Hannover). Werz employed domino reactions based on donor-acceptor-substituted cyclopropanes to build up di- and oligomers of various five-membered heterocycles. The reported rearrangement cascade leads to (multiply) 3,3'-linked products. Gaich showed how a Cope-type rearrangement may enable carbon-carbon bond formation on the least nucleophilic 4-position of indoles, mimicking the key step in the proposed biosynthetic pathway of alkaloids by dimethylallyltryptophan synthase. The talks of Leandro Andrade (University of São Paulo) and Rebecca Goss (University of St. Andrews) provided a thematic link to the following, more biologically orientated sessions. Andrade studies enzymatic reactions of organic substrates containing heteroatoms like selenium, boron, and silicon. In addition to this work, he also described his efforts to isolate previously unknown bacterial strains from different environments, including the Antarctic peninsula, providing fascinating details on the sample acquisition in the latter habitat. Goss introduced *genochemetics*, that is the process of introducing a gene into an organism, which works in concert with an existing biosynthetic pathway of a natural product. By this method, a chemical handle is installed on the periphery of the natural product, which allows for subsequent diversification of the parent compound by chemical means.

### Breakout

The meeting also included a breakout session, allowing the participants to discuss three pre-selected topics associated with the running of a research group. These topics were the role of basic research in today's science, the challenges of running and organising a group, and the role of industrial partnerships. Each group's conclusions were presented to the delegates in a subsequent joint discussion. With respect to basic research, the common concern was that the balance

in research funding has shifted in recent years too much towards more industry/application-oriented projects; this was also the theme of Prof. Dr. Helmut Schwarz's after dinner lecture. Several participants suggested that the national scientific societies should be doing more to lobby funding councils and politicians to stem the flow away from basic research, and that researchers need to more actively promote the fundamental importance of basic research (through outreach activities) to the general public. The discussion on research group organisation concluded with most participants agreeing that the step from a successful postdoctoral fellow to a principal investigator (PI) is very large, and universities need to provide far more training to early-career academics to better prepare them for the challenges involved. With regard to industrial partnerships, three main challenges were identified in the breakout section. These include intellectual property issues (patents versus publications), the difficulty of finding industrial partners (especially at an early stage of one's career), and difficulties in identifying mutually relevant and interesting research topics.

### Natural Product Chemistry

The chemical biology sessions were an excellent representation of the diversity and multidisciplinary nature of this exciting field. Talks ranged from the development of new bio-conjugation methods for the labeling of proteins to the use of chemical tools to understanding critical processes in cancer.

The session on natural products focused on the discovery of novel, bioactive structures and scaffolds from a diverse variety of bacteria and fungi. The natural world remains a fertile ground for the discovery of potential therapeutics, and several speakers detailed their efforts to uncover such molecules. Hendrik Luesch (University of Florida) detailed his group's efforts in identifying an antimicrotubule agent from marine cyanobacteria. Their efforts, including the total synthesis of the parent molecule and several derivatives, as well as examining the biological activities of the compounds opened this session. Another interesting use for natural products was outlined by Hans-Dieter Arndt (Friedrich-Schiller-Universität Jena). His group has utilized a fluorescently labeled derivative of a compound that binds actin as an imaging tool. As natural products typically bind their targets with high affinity and selectivity this may be a useful approach to utilizing such molecules as imaging tools.

In order to manipulate living systems for the production of natural products and derivatives one must first understand the pathways used to synthesize them. Several speakers in this session outlined their groups' efforts to understand the pathways used to make a variety of naturally occurring molecules in several organisms. These compounds ranged from terpenes to non-ribosomal peptides. It was clear that the challenge of unraveling these pathways is significant, howev-





Figure 5. The conference participants on the excursion to the Kampenwand mountain in Aschau; photo: A. Wirt and H.-G. Weinig (GDCh).

er, there is much potential in manipulating naturally occurring pathways for the production of bioactive molecules.

### Excursion into the Bavarian Countryside

Despite the early morning starts, and late night finishes, the conference was not all work, work, work. On the afternoon of the penultimate day, the participants enjoyed a hike up the beautiful Kampenwand mountain in Aschau (Figure 5). The scenery was breathtaking, as were the sheer



Figure 6. Putting theory into practice: A few of the first to arrive at the summit: left to right: David Glowacki, Timo Jacob, Thomas Miller III, Anastassia Alexandrova, Alexandre Tkatchenko, Lasse Jensen, Birgit Strodel; photo: A. Alexandrova.

drops either side of the path taken by some to the summit (Figure 6). The chairman was particularly keen to ensure that we did not lose any of the participants off the side of the mountain! This was followed by a traditional Bavarian dinner at an “Alm-style” restaurant with beautiful views of the sun setting behind the mountains. Once back at the monastery, several of the delegates decided to go for a late night swim in the lake. The next day, participants reported warm midsummer night weather, the cool lake waters and excellent views of the Milky Way, and to top it all, excellent views of the Perseid meteor shower. As one delegate put it: “it was so beautiful and romantic, we were in the lake looking up and could clearly see the Milky Way, and then the meteors came. It was just a real shame that I was with a bunch of scientists and not my wife...”

### Chemical Biology

The theme of understanding natural product biosynthesis pathways continued into the next session, titled “Tools in Chemical Biology” by Manuela Tosin (Warwick University). This and the following session titled “Methods and Processes in Chemical Biology” offered a good representation of the increasing diversity of chemical biology research, and the future direction of this field. It is evident that a large number of chemists are using the wide variety of molecules, tools and techniques available to them to probe and understand biological pathways, networks, and systems. Approaches such as metabolomics and chemoproteomics have (and continue to) significantly increase our understanding of the metabolic changes associated with cancer and have the potential to fundamentally alter our approach to treating this disease. Daniel Nomura (University of California, Berkeley) detailed the use of such a platform for the discovery and characterization of the specificity of inhibitors that

target the same class of proteins. Another potentially significant approach to treating disease is to target the protein–protein interaction of transcription factors that are activated in cancer. Ali Tavassoli (University of Southampton) detailed discovery and validation of cyclic peptide inhibitors of these “undruggable” targets. Using engineered bacteria, libraries of around 100 million cyclic peptides were screened for inhibitors of two transcription factors critical to tumor formation and growth. These molecules were said to not only form the starting point of novel therapeutics, but also serve as important biological tools that allow better understanding of cancer biology. Another speaker using high-throughput screening to discover molecules that regulate the cell cycle was Ulrike Eggert (King’s College London), whose lab are working to uncover and characterize a series of molecules that target various proteins and pathways in cytokinesis, for use as a molecular toolbox to allow better understanding of cell division and cell cycle regulation. It was evident from both these talks that high-throughput screening approaches continue to play a critical role in chemical biology, and the discovery of new bioactive molecules.

Several speakers detailed the development and use of new molecules for labeling proteins in cells. Christian Hackenberger (Humboldt Universität Berlin) has taken the use of click chemistry a step further by introducing Staudinger to Huisgen; using a linker capable of undergoing a stepwise Staudinger and CuAAC reaction opens the possibility of

two-step, chemoselective labeling reactions. This and several other talks demonstrated the significant advances being made in the protein-labeling field, with the detailed mechanistic understanding of chemists enabling the development of novel labeling reagents for use in imaging biological systems. Classic chemical reactions are being continually adapted for use as novel bioorthogonal reactions (as detailed by Neal Devaraj from the University of California, San Diego) with biomolecules such as DNA and amino acids. The field of nucleic acid chemistry was also represented in these sessions. Glenn Burley (Strathclyde University) detailed several molecules that regulate RNA splicing, thus enabling his group to gain insight into the labyrinth of the spliceosome, while Danica Fujimori (University of California, San Francisco) outlined her group’s efforts to unravel the mechanism of two recently discovered RNA and DNA methylating enzymes.

Overall the meeting was an excellent symposium that enabled international and multidisciplinary networking opportunities for the young guns of chemistry in the UK, USA, Germany, and Brazil. Unlike subject-specific meetings, the TFOC Symposium exposed its participants to the breadth and diversity of current chemical research, an approach that will hopefully enable new, sustainable transdisciplinary collaborations.



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Dr. Stefan M. Huber



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