

Editorial: The Future of Chemical Physics Conference 2016

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Editorial: The Future of Chemical Physics Conference 2016

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I. INTRODUCTION

The first conference organized and sponsored by *The Journal of Chemical Physics* (JCP) took place in Oxford, UK at St. Edmund Hall and the University of Oxford Physical and Theoretical Chemistry Laboratory on August 31st through September 2nd, 2016. The conference, entitled “Future of Chemical Physics,” was part of the AIP Publishing Horizons Conference Series. The JCP Editors Angelos Michaelides, David E. Manolopoulos, Carlos Vega, Peter Hamm, and Marsha I. Lester organized the scientific program and served as session chairs, along with JCP Editor David W. Chandler and several European JCP Editorial Advisory Board (EAB) members. Several other EAB members attended the conference.

The program aimed to provide an overview of the most exciting recent developments in chemical physics and explore where the field is headed in the next 10–20 years. The conference was very broad with three presentations in each of the Table of Contents sections of the journal. The 21 invited speakers were asked to provide an overview of their own subfield, accessible to a broad audience, and also to conclude their talks with a slide on future challenges.

This editorial is intended to share the ideas discussed at the conference with the broader chemical physics community. Highlights of the 21 invited presentations follow, in addition to a group photograph (Figure 1) and information about the poster prize winners. The program book is included in the [supplementary material](#).

II. ATOMS, MOLECULES, AND CLUSTERS

Shedding IR Light on Gas-Phase Metal Clusters: Insights into Structures and Reactions

André Fielicke (Technische Universität Berlin)

André Fielicke spoke about work aimed at understanding clusters and aggregates of atoms and small molecules. He pointed out that many of the fundamental tasks of science, such as ensuring efficient food production, clean energy usage, and minimizing the environmental impact of our lives, rely on understanding reactivity at surfaces and in particular

catalytic behavior at surfaces. Much of this research has relied on experiments that look at bulk materials, but new techniques that utilize sensitive action spectroscopies allow observation of the structure and reactivity of size and charge selected nano-materials. With techniques such as infrared photon-induced ionization or dissociation of rare-gas-tagged clusters, it is now possible to obtain IR spectra of small reactive clusters. These data are important in their own right, but also as a stringent test of the accuracy of modern electronic structure theories. As these techniques evolve and become more accessible, the opportunity exists to perform time-resolved monitoring of catalytic clusters as they react. The more specificity and detailed knowledge that is obtained, the more one will be able to design efficient catalysts from readily available materials.

Challenges in the Chemical Physics of Aerosols

Jonathan Reid (University of Bristol)

Jonathan Reid spoke of the tremendous new knowledge that is being developed of reactivity and properties of aerosols and droplets that are so important to the understanding of our environment and in a wide range of applications. The combination of surface effects and highly supersaturated bulk concentrations of species, both neutral and ionic, throughout a droplet makes understanding these systems in detail a very complex and interesting chemical physics problem. Studies on single droplets allow focus on investigations into controlled growth, evaporation, and crystallization of aerosols. Changes in aerosol properties, such as surface tension, surface charge, concentration gradients, and alignment and orientation of molecules at the surface, impact the reactivity, morphology, and optical properties of the aerosols. New spectroscopic techniques are being developed to study these species, and this will remain a challenging area for chemical physics for many years as the nature, applications, and impacts of aerosols change.

State-of-the-Art Imaging Techniques for Chemical Dynamics Studies

Claire Vallance (University of Oxford)

Claire Vallance highlighted new tools being used to study the photochemical properties of single molecules. In



FIG. 1. Future of Chemical Physics 2016 group photograph.

particular, the focus was on larger molecules with many complex pathways for photochemistry. These complex systems require novel experimental tools, such as new multi-dimensional imaging techniques for the detection of many products from a single molecular photo-dissociation event. New cameras are being developed that allow each pixel to record multiple time-stamped events before the pixel array is read out. This allows one to measure the position and arrival time of multiple ionized species from a single photo-dissociation event. These new cameras are finding uses in multi-mass covariance-map imaging of Coulomb explosions and multiplexed mass spectrometry. Beyond single molecules in the gas phase, Vallance explored opportunities for looking at molecules at surfaces and in membranes, with future developments in these techniques potentially impacting a large community of researchers.

III. LIQUIDS, GLASSES, AND CRYSTALS

Facets of Glass Physics

Ludovic Berthier (Université de Montpellier)

Ludovic Berthier spoke on the glass transition that occurs when liquids are cooled quickly. It is often possible to avoid the formation of a crystalline phase and to obtain a glass that is dynamically arrested, presenting high viscosity but not long range positional order. The change in viscosity of a liquid with temperature is quite abrupt and denoted as the glass transition. He pointed out that understanding this transition from a theoretical point of view will continue to be an important area of research. Ideally one could produce better glasses by slower cooling. Recently discovered vapor deposited glasses produce non-conventional ultra-stable glasses, and this seems another

promising route. The aim is to obtain experimental systems that come closer and closer to the theoretical concept of the ideal glass transition and reveal a direct signature of the microscopic mechanisms at play. He also indicated that it would be desirable to find algorithms that produce better equilibrated glasses in computer simulations, maybe with moves that, although unphysical (for instance, swap moves that exchange the positions of two types of particles in mixtures), allow one to get much closer to the ideal glass transition or to dispel its relevance.

Is the Glass Transition Universal?

Kristine Niss (Roskilde University)

Kristine Niss described how experimentalists have tried to establish universal behaviors and correlations in an effort to guide the development of a theory for the glass transition. However, she pointed out that when more systems are studied, predictions are usually found to hold only for a limited class of systems. The emerging picture is that while the glass transition can be observed in many systems independent of chemical details, there is also a myriad of specific behaviors and it seems unlikely that this can all be captured in one theory. Based on this understanding, it is relevant to address the question: what are the features that should be included in the “ideal gas model” of glass forming liquids? By establishing a framework for understanding the glass transition in its simplest version, we will gain the best starting point to understand the role of the glass transition in complex liquids, soft and biological matter in the future. In addition, an interesting line of research for the future is to understand what controls the competition between crystallization and glass formation.

*Aqueous Nanoscopic Systems***Sylvie Roke (École Polytechnique Fédérale de Lausanne)**

Sylvie Roke illustrated how the study of aqueous interfaces is of relevance to geo, biological, and energy sciences. She presented a number of emerging spectroscopic techniques that can be used to understand the ordering and composition of molecules at interfaces on a nanoscopic and microscopic basis, such as sum frequency scattering (SFS) and second harmonic scattering (SHS). In the future, these techniques will help understand both the structure and the dynamics of molecules at the interface as well as its composition (for instance, clarifying the acidic or basic nature of the water surface, or the effect of ions on the surface of water at high dilution, and the significant changes that occur when deuterated water is used as a solvent). Understanding the interface between water and hydrophobic liquids is also of relevance to understanding hydrophobicity and the mechanism behind the formation of nano-emulsions, micelles, and biological membranes. She also indicated that an important future area of research is to map spatially and temporally resolved interfacial changes using femtosecond second harmonic and sum frequency microscopes. The molecular level understanding of aqueous interfaces, obtained from spectroscopic measurements, will eventually help map changes in living systems.

IV. SURFACES, INTERFACES, AND MATERIALS*Ionic Liquids—A Challenge to Our Understanding of the Liquid State***Susan Perkin (University of Oxford)**

Susan Perkin started with a historical perspective and broad overview of contemporary understanding of ionic liquids, particularly ionic liquids at surfaces. She talked, amongst other things, about recent surface force measurements on electrolyte solutions from her lab, which showed surprisingly large long-range electrostatic interactions. It was particularly apparent from Perkin's talk that in the "field" of ionic liquids, experiment, theory, and computer simulation are all needed if genuine fundamental understanding of these complex solutions is to be obtained. Some of the challenges and opportunities identified were the need for theories capable of understanding and describing ionic liquids, plasmas, concentrated electrolytes, and molten salts on the same footing. The relationship between nanostructure and physical properties was also identified as a challenge for our understanding of the chemical physics of ionic liquids.

*Uncovering the Photophysics of Organic and Hybrid Semiconductors***Natalie Banerji (University of Fribourg)**

Natalie Banerji gave a talk on a wide variety of organic, inorganic, and hybrid semiconductors and the fundamental insight obtained on these from techniques such as ultrafast spectroscopy. This included work from her group on charge generation in organic solar cells, charge transport in polyelectrolyte films, and on the prospects of interfacing organic semiconductors with biology. For the last topic, it was emphasized that fundamental chemical physics insight

is required to speed up the development of future organic-bio-electronic devices, which have many exciting applications, e.g., in health care. Key fundamental questions identified for the field included the need to better understand the interaction of light with matter and charge transport and dynamics, particularly at complex "real world" interfaces. The development of tools for ultrafast and terahertz spectroscopy on real operating devices will certainly help in addressing these issues.

*Disorder, Defects, and Temperature: Modelling Real Materials from First-Principles***Aron Walsh (Imperial College London)**

Aron Walsh gave a talk on modelling materials with *ab initio* electronic structure approaches, with a particular emphasis on imperfect crystals. He reported work being carried out in his group on hybrid halide perovskites (an interesting class of materials for solar energy) and emphasized the importance of treating thermal effects and long-range disorder for a proper microscopic description on these materials. Looking forward, Walsh talked about various challenges and opportunities the field faces in moving beyond the treatment of static defect-free crystals, which has been the focus of the vast majority of electronic structure studies of solids. In passing he noted that improvements in algorithms, computer software, and hardware mean that the field of computational materials science is now data rich. This concept of "big data" presents a specific set of challenges and opportunities for the field (associated with sharing, curation, and mining of data) as expanded upon later in the meeting (see Sec. VIII).

V. POLYMERS AND SOFT MATTER*Directed Self-Assembly of Janus Rods***Dirk Aarts (University of Oxford)**

Dirk Aarts discussed how in recent years it has become possible to synthesize colloidal particles with specific interactions and well defined shapes in relatively large quantities. The interesting feature is that the interactions between the colloidal particles can be "tuned" in the lab. He illustrated how this offers a unique opportunity to obtain systems with tailored properties, from both a thermodynamic and a dynamic point of view. Besides this, developments in experimental techniques such as microscopy, soft-lithography, and laser tweezing now allow us to extract quantitative information down to the particle level in 3D, fully time resolved. He concluded that this area of soft condensed matter offers exciting opportunities in the future for a broad community of researchers working on experimental techniques such as synthesis and imaging and in theory and simulations.

*Multiscale Modeling of Soft Functional Materials Based on Responsive Polymers: Towards Adaptivity and Feedback Control***Joachim Dzubiella (Helmholtz-Zentrum Berlin)**

Joachim Dzubiella described how metallic nanoparticles can now be used for catalysis, often described as nanocatalysis. However, in the case of metallic nanoparticles and poly-

meric colloidal particles, it is frequently necessary to cover the particle with a grafted polymer to avoid the aggregation of the particles. He discussed how the properties of these grafted polymers change significantly with temperature, pH, and other variables so that the shell covering the particles is highly responsive to changes in the system. Properties like transport (from the bulk to the particle) and solute distribution will change significantly when the polymer shell covering the particle changes its properties. This is a complex problem with a spatiotemporal coupling of transport and particle distribution over many scales. Ideally one would like to understand these processes and also perform a rational design of these particle-polymer shell materials so that they have the desired programmable response. The area of soft functional polymer materials (including nanocatalysis) is a promising area of research where it is crucial to understand the non-equilibrium phenomena that arise and how the complex problem can be mapped into a simpler “coarse grained” picture.

Surface Design with Polymers

Friederike Schmid (Johannes Gutenberg Universität Mainz)

Friederike Schmid discussed the brushes formed when polymers are tethered to a surface. The properties of these grafted chains can be different from those in the bulk. These brushes can be used to stabilize colloids. However, they also play an important role in friction and problems related to nanotribology. Brushes can be used to control the properties of the surface to which they are grafted. In her talk, she showed how it is possible to design them such that they respond sensitively to changes in the thermodynamic and environmental conditions, e.g., by mixing different types of grafted polymers or by attaching active groups that can switch from being exposed to buried inside the brush. By playing with the thermodynamic conditions, one could develop active switches on the nanoscale, for instance, changing the surface from hydrophilic to hydrophobic. She pointed out that it might also be possible to design surfaces with switchable slip. She concluded by mentioning that brushes can also be used in catalysis, and it would be of interest to be able to move from designing surface properties to designing processes at surfaces.

VI. BIOLOGICAL MOLECULES AND NETWORKS

Biomolecular Hydration Shells: Dynamics and Biochemical Function

Damien Laage (École Normale Supérieure)

Damien Laage addressed the question of the extent to which water affects the function of bio-macromolecules, for example, in the context of crowding effects in a cell or enzyme catalysis. In his view, water is important for the dynamics and function of a protein, but “biological water” is quite similar to bulk water, even though a consensus on this point has not been reached yet. As a long-term goal, a better understanding of the role of water might be a step towards the question whether or not other forms of life are conceivable in solvents other than water.

Biophysics at the Nanoscale: Single-Molecule Spectroscopy of Protein Folding and Dynamics

Ben Schuler (University of Zürich)

Ben Schuler’s research is concerned with the dynamics of proteins measured by single molecule spectroscopy, trying to push the time resolution of the method to its limits by combining Förster resonance energy transfer (FRET) with sophisticated correlation spectroscopy and a quantitative analysis of the statistics of the fluorescence photons. He sees the biggest challenges in a more integrated approach of experiment, computer simulation, and analytical theory, which will be needed to ultimately develop a physical picture of protein dynamics and function. Indeed, the value of combining experiment, simulation, and analytical theory was a recurring theme of the meeting.

Advances in the Simulation of Protein Aggregation at the Atomistic Scale

Birgit Strodel (Forschungszentrum Jülich)

Birgit Strodel studies the physicochemical principles that govern the highly complex process of protein aggregation from a computer simulation point of view. In particular, she addressed the length- and timescale problems of computer simulations, which to date often cover orders of magnitude smaller scales than what is probed experimentally. Other issues concern the concentration problem when studying protein aggregation, since molecular dynamics simulations are typically done at unrealistically high protein concentrations, as well as the development of transferable force fields, which are not tailored to the folded state of a protein.

VII. ADVANCED EXPERIMENTAL TECHNIQUES

Investigating Structure Using Solid-State NMR: Principles, State-of-the-Art, and Challenges

Sharon Ashbrook (University of St. Andrews)

NMR spectroscopy is undoubtedly one of the most powerful spectroscopic techniques to investigate the structure and dynamics of small to mid-sized molecules, but becomes extremely challenging when solids or materials are investigated. The research of Sharon Ashbrook is concerned with developing solid-state NMR for these types of samples to the same level as conventional NMR for small organic molecules. The challenges lie in both the low spectral resolution and sensitivity of solid-state NMR, which may be addressed by higher magnetic fields, ultrahigh speed magic angle spinning (MAS), and dynamic nuclear polarization (DNP). In her talk, Ashbrook showed a number of interesting examples which demonstrated the power of solid state NMR to problems in materials science, particularly when used in conjunction with complementary electronic structure calculations.

Pushing Chemical Physics to the Nanoscale

Niek F. van Hulst (ICFO–The Institute of Photonic Sciences)

Niek F. van Hulst aims to track the dynamics of molecules in action in real-life complex (bio)environments. His work tries to bridge ultrafast and ultra-small, i.e., he designs experiments

that combine single molecule detection with the time-resolution needed to resolve even the fastest chemical processes occurring on typically a 100 fs time scale. One approach to achieve this goal is to combine pump-dump experiments with single-molecule fluorescence detection, while the future dream experiment appears to be single-molecule 2D spectroscopy.

Visualising Nanoscale Structure and Dynamics Using “Chemical Physics”

Philipp Kukura (University of Oxford)

Philipp Kukura talked about single molecule detection without fluorescence by using merely the contrast generated by a different index of refraction of, e.g., a protein versus that of the solvent. This approach allows one to study non-fluorescing molecules and as such avoids having to label them. A sensitivity down to the single protein level has been demonstrated, which paves the way for a wide variety of applications in biosensing and for investigations of protein-protein and protein-substrate interactions.

VIII. THEORETICAL METHODS AND ALGORITHMS

Using Machine Learning to Map the Structure and Predict the Properties of Materials and Molecules

Michele Ceriotti (École Polytechnique Fédérale de Lausanne)

Michele Ceriotti discussed applications of machine learning techniques to problems in chemical physics. His talk covered both the fitting of *ab initio* potential energies to simple functional forms based on the local environment of each atom, and the automatic generation of appropriate collective variables for constructing and visualizing the free energy landscapes of proteins and complex materials. Machine learning techniques are being developed at a rapid pace by software companies who are interested in organizing and interpreting large amounts of data. This talk made it clear that these developments can have a tremendous impact in the realm of theoretical chemistry, provided that an effort is made to formulate a representation of molecular structure that encodes chemical insight and physical symmetries. Applications of machine learning techniques to chemical physics are still relatively new, but are expected to increase as the field moves towards the study of larger and more complex systems.

Quantum Electrodynamics of the Early Stages of Photosynthetic Light Absorption

Fred Manby (University of Bristol)

Fred Manby began by presenting an overview of modern electronic structure theory, in which he described both density functional theory and more accurate wave function methods, and how it is possible to “embed” the latter in the former (i.e., to treat a subsystem of interest with a higher level of electronic structure theory than the surrounding bath). He then went on to describe some recent work from his group on how to set up the correct initial state for photosynthetic excitation energy transfer under low light intensity,



FIG. 2. The poster prize winners with Editor-in-Chief Marsha I. Lester in the lobby of the Physical and Theoretical Chemistry Laboratory at the University of Oxford. From right to left: Back row: Yixing Chen, Christopher Hall, Hamish Hiscock, Susannah Worster, and Rebecca Ingle. Front row: Nikolay Smolentsev, Robert Baldock, and Marsha I. Lester.

which he illustrated with a model simulation of the Fenna-Matthews-Olson complex with parameters obtained from electronic structure calculations. The future challenges identified included harnessing the power of modern massively parallel computer architectures and unifying electronic structure theory with dynamics. He also welcomed the advances in machine learning and was optimistic about opportunities for stronger interactions between machine-learning and quantum chemistry.

New Directions for Monte Carlo Algorithms in Electronic Structure Theory: FCI, CASSCF, and Multi-Reference Perturbation Theory

Ali Alavi (University of Cambridge/MPI for Solid State Research)

Ali Alavi reviewed the development of his “full configuration interaction quantum Monte Carlo” (FCIQMC) algorithm from its birth in *The Journal of Chemical Physics* seven years ago to the stage where it is now proving to be a powerful tool for shedding light on strongly correlated electrons. Many examples of this were given in the talk, including application to the electronic structure of cuprates, in which the FCIQMC algorithm was used to speed up the configuration interaction step in the construction of a complete active space self-consistent field (CASSCF) wave function. Looking ahead, Alavi emphasized the general challenges for electronic structure theories in treating strong multi-configurational systems and in combining high-level electron correlation methods with nuclear optimization and dynamics.

IX. CONCLUSIONS

The conference concluded with a panel discussion on the *Future of Chemical Physics* led by David Manolopoulos with panelists Jeppe Dyre (Roskilde University), Thomas Elsaesser (Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie), James T. (Casey) Hynes (University of Colorado

Boulder, École Normale Supérieure), David H. Parker (Radboud University), and Michele Parrinello (ETH Zürich). While it is difficult to make predictions about the future, the invited talks, poster sessions, and concluding panel discussion revealed many active and emerging areas of chemical physics research that are ripe for future exploration.

Five of 79 posters were awarded poster prizes. The poster prize winners (pictured in Figure 2) are as follows:

Nikolay Smolentsev and **Yixing Chen** (École Polytechnique Fédérale de Lausanne) co-presenting “Intermolecular Headgroup Interaction and Hydration as Driving Forces for Lipid Transmembrane Asymmetry/Electrolytes induce long-range orientational order and free energy changes in the H-bond network of bulk water”

Rebecca Ingle (University of Bristol) presenting “Structure or Dynamics: What Drives Photoproduct Branching Ratios at Conical Intersections?”

Susannah Worster and **Hamish Hiscock** (University of Oxford) co-presenting “Long-lived spin coherence in the avian magnetic compass”

Christopher Hall (University of East Anglia) presenting “Ultrafast isomerization dynamics of a unidirectional

molecular rotor revealed by femtosecond stimulated Raman spectroscopy (FSRS)”

Robert Baldock (University of Cambridge) presenting “Hamiltonian Monte Carlo nested sampling”

SUPPLEMENTARY MATERIAL

See [supplementary material](#) for the program book for the Future of Chemical Physics Conference.

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