CPP 49: Soft Matter Physics: Emerging Topics, New Instruments and Methods

Time: Wednesday 15:00–17:45   Location: C 230

CPP 49.1   Wed 15:00   C 230
Collective Hydration Dynamics in Binary Mixtures: A THz Time Domain Spectroscopic Study — •Debasish Das Mahanta, Dipak Kumar Das, Animesh Patra, Nirnay Samanta, and Rajib Kumar Mitra — S. N. Bose National Centre for Basic Sciences, Block-JD, Sector-III, Saltlake, Kolkata-700106

We have studied the structure and dynamics of water in its binary mixture with two amphiphilic molecules 1,2-dimethoxy ethane (DME) and dimethyl sulfoxide (DMSO) by THz time domain spectroscopy (TTDS) (0.3-1.6 THz region). In both the cases a non-ideal behavior of the mixture is found owing to the formation of water clusters. The cooperative dynamics of water in those binary mixtures, obtained from Debye relaxation of TTDS data reveals a non-monotonous behavior as a function of water concentration (X_w).

CPP 49.2   Wed 15:15   C 230
Initiation of bimolecular ground-state reaction via IR excitation — •Till Stensitzki[1], Yang Yang[1], Valeri Kozich[1], Ashour A. Ahmed[2,3], Florian Kössel[1], Oliver Kühn[3], and Karsten Heyne[1] — [1]Department of Physics, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany — [2]Faculty of Science, Department of Chemistry, University of Cairo, 12613 Giza, Egyp — [3]Institute of Physics, University of Rostock, Albert Einstein-Strasse 23-24, 18059 Rostock, German

Excitation of vibrations that participate in the reaction coordinate are believed to accelerate an otherwise thermally driven chemical reaction. Here, we demonstrate the acceleration the thermal driven chemical reaction between cyclohexanol and phenylisocyanate in THF. Upon excitation of the OH-stretching vibration of CH-ol we track bleaching signals of PHI and the urethane product band in real-time. Additionally, we determined reaction rate changes upon selective vibrational excitation with negligible heating of the sample and observed an increase of the reaction rate up to 24%. We utilized reaction acceleration between toluene-2,4-diisocyanate and 2,2,2-trichloroethane-1,1-diol to write a polyurethane square on the sample windows by femtosecond IR pulses.

CPP 49.3   Wed 15:30   C 230
New neutron spectrometer NEAT at Helmholtz Zentrum Berlin for investigation of functional dynamics at nanoscale — Gerrit Günther, Veronika Grzimek, and •Margarita Russina — Helmholtz Zentrum Berlin für Materialied und Energie, Hahn-Meitner Platz 1, 14109 Berlin

Neutron time-of-flight spectrometer NEAT has a long history of successful application to the study dynamics and function. It is best suited to probe dynamic phenomena directly in the broad time domain of 0,1-100 ps and on the large length scale ranging from 0.05 to up to about 5 nm. Recent upgrade of the instrument resulted in 300 fold higher count rate compare to predecessor placing NEAT on the level of the world best. The advanced features of the new instrument include novel integrated guide-chopper system which deliver neutron beam with variable characteristics and substantial increase of the detector angle coverage with simultaneous increase of angular resolution. New instrumental capabilities provides optimal experimental conditions for investigation of microscopic dynamics in soft and hard matter including single excitations in crystals, applications with high magnetic field up to 14 T or in-situ confinement of gasses and liquids in porous systems. Here we present details of NEAT upgrade, measured instrument characteristics and show first experimental results.

CPP 49.4   Wed 15:45   C 230
Second-Harmonic Scattering: Theory and Atomistic Simulation — •David M. Wilkins[1], Sylvie Roke[2], and Michele Ceriotti[1] — [1]Laboratory of Computational Science and Modeling, IMX, EPFL, Lausanne, Switzerland — [2]Laboratory for fundamental BioPhotonics, Institutes of Bioengineering and Materials Science and Engineering, School of Engineering, and Lausanne Centre for Ultrafast Science, EPFL, Lausanne, Switzerland

Recent second-harmonic scattering (SHS) experiments on electrolyte solutions have shown evidence of long-ranged ion-induced intermolecular correlations between solvent molecules, persisting on the ∼10 nm length scale at sub-molar concentrations. SHS is exquisitely sensitive to these subtle structural changes, and an atomistic understanding of the kinds of correlations probed is desirable.

I discuss the development of a computational framework for modelling SHS experiments of general condensed-phase systems. This includes two main ingredients: firstly, the orientational correlations between molecules, and secondly, the molecular hyperpolarizability tensor. I show that, at variance with common assumptions, even in an isotropic liquid the modelling of the SHS signal requires that correlations be accounted for, and that the hyperpolarizability tensor depends strongly on the molecular environment. Further, I discuss a machine-learning framework we have developed to sidestep computationally expensive calculations of this tensor, which provides a large step towards the routine calculation of SHS intensities of general systems using machine-learning methods.

CPP 49.5   Wed 16:00   C 230
Symmetry-Adapted Machine-Learning for Tensorial Properties of Atomistic Systems — •Andrea Grisafi, David Mark Wilkins, and Michele Ceriotti — Laboratory of Computational Science and Modeling, EPFL, Lausanne, Switzerland

In the last few years, machine-learning methods have played a prominent role in providing an accurate description of atomic-scale properties, bypassing computationally demanding quantum chemistry calculations.

A great deal of effort has been applied to learning the ab initio ground state energy of materials and molecules, giving accurate potential energy surfaces. However, a complete description of a condensed phase system requires the calculation of properties that are tensorial in nature, e.g., multipole moments. Unlike scalar quantities, these properties transform in prescribed ways when the reference frame is rotated, presenting a great challenge when designing a machine-learning algorithm.

I discuss an extension of a traditional machine-learning method which can be used to predict tensorial properties of arbitrary rank for systems of arbitrary complexity, automatically encoding the rotational symmetry in three dimension. The novel algorithm has the potential to cover a broad range of applications within material science and condensed matter physics. Applications are shown for the prediction of the electrical response series of water oligomers, from the single molecule to the condensed-phase. Preliminary results for the prediction of the ab initio charge density of organic molecules will be also presented.

CPP 49.6   Wed 16:15   C 230
A generalized convex hull construction for computational materials discovery — •Andrea Anelli, Edgar Engel, and Michele Ceriotti — EPFL, Lausanne,Switzerland

Computational materials searches have the ultimate goal of discovering technologically relevant materials with desired physical properties. State of the art methods can generate thousands of locally-stable configurations. Selecting among these the few structures that can be experimentally synthesised is a needle-in-a-haystack kind of problem.

Conventionally, this screening is performed using a convex hull construction (CH), which can only identify structures stabilized by manipulation of a particular thermodynamic constraint (e.g. density or composition) chosen on the basis of experimental evidence or intuition.

The predictive power of such a formulation is weakened by its dependence on prior knowledge of the system, disregarding configurations that could appear as (meta-)stable under unconventional choices of constraints. We introduce a generalized CH based on an abstract representation of structural features, which are dependent solely on the cartesian coordinates of the systems and constitute a general, agnostic set of thermodynamic constraints. We further introduce a probabilistic scheme that is consistent with uncertainty quantification of the convex-hull construction, reducing the impact of modelling errors on the determination of synthesizable structures. We demonstrate the predictive power of our formulation by applying it to Hydrogen at TPa pressures, the binary high-pressure Oxygen-Hydrogen system and, increasing the structural complexity of the system, on Pentacene molecular crystals

15 min. break

CPP 49.7   Wed 16:45   C 230

In-situ characterization of NiFeOOH for the oxygen evolution reaction using X-ray absorption spectroscopy — •Dorian Drevon — Albert-Einstein-Strasse 15, Berlin

Reducing global carbon emissions will require efficient catalysts for use in solar-to-fuel conversion processes, where electrochemical water oxidation is the key to a zero emission cycle. For this, understanding the fundamental processes of the oxygen evolution reaction (OER, 4OH- * O2 + 2H2O + 4e-) from a mechanistic perspective is an important goal for the design of highly efficient and functional electrocatalysts.

The Ni-Fe oxyhydroxide electrocatalysts are currently the most active OER catalysts in alkaline media Combining Ni and Fe leads to an excessive increase in the catalytic activity and a coincident change at the metal redox states, witnessing of the electronic interaction between Ni and Fe sites. However, the complexity of the metal redox activity have resulted in a debate regarding the OER active state in this electrocatalyst.

In-situ O K-edge X-ray and Fe,Ni L-edge absorption fine structure (XAFS) spectroscopy was used to investigate the electronic and structural change in the Nickel Iron electrocatalyst during the oxygen evolution reaction (OER). Above 0.5V versus Ag/AgCl, we were able to see the rise of pre-features at the Oxygen k-edge and post feature at the Nickel L-edge but they were absent in the Fe L-edge suggesting that Ni undergoes a change in its oxidation state from Ni(II) to of Ni(III,IV) when the catalysts becomes active.

CPP 49.8   Wed 17:00   C 230

In-situ R-SoXS study of the structure formation during solvent annealing in organic thin films for application in photovoltaics — •Mihael Coric[1], Isvar Cordova[2], Gregory Su[2], Feng Liu[3], Cheng Wang[2], and Eva M. Herzig[1,4] — [1]TU München, Munich School of Engineering, Herzig Group, 85748 Garching, Germany — [2]Advanced Light Source, Lawrence Berkeley National Laboratory, 94720 Berkeley, USA — [3]Department of Physics and Astronomy, Shanghai Jiaotong University, 200240 Shanghai, China — [4]Universität Bayreuth, Physikalisches Institut, Herzig Group - Dynamik und Strukturbildung, 95440 Bayreuth, Germany

Resonant X-ray scattering (R-SoXS) at the carbon edge is now an established, very powerful tool for the analysis of structure and morphology formation in organic thin films. Since soft X-rays have a very low penetration depth and strong air scattering, samples need to be measured in high vacuum, but offer a much higher contrast than hard X-rays. However, in regards to the examination of structure formation, in-situ studies are essential to achieve a better understanding of the formation processes taking place in thin films. In this work, we present a method using the well-known R-SoXS technology with a setup that enables us to execute in-situ studies under a steady flow of vapor inside the vacuum chamber. We will show that it is possible to track changes in the morphology of organic thin film materials during a solvent annealing process. In combination with in-situ UV-Vis measurements, we are able to gain a better understanding of the structure and formation processes involved.

CPP 49.9   Wed 17:15   C 230

EMIL goes operando: _In-situ_ characterization of real-world electrocatalysts by soft x-ray spectroscopy — •Raul Garcia-Diez[1], Regan G. Wilks[1,2], and Marcus Bär[1,2] — [1]Renewable Energy, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH (HZB), Berlin (Germany) — [2]Energy Materials In-Situ Laboratory Berlin (EMIL), HZB, Berlin (Germany)

The _in-situ_ study of catalyst materials during the electrolysis of water is of vital importance to understand the limiting mechanisms occurring in the catalytic reaction. The combination of soft x-ray absorption (XAS) and emission (XES) spectroscopies is an established method that can probe the chemical and electronic structure of the electrocatalyst and give insights into its local density of states and, therefore, its catalytic behavior.

For this purpose, in the Energy Materials In-Situ Laboratory Berlin (EMIL) at the BESSY II synchrotron facility we are developing an infrastructure that allows XAS, XES, and RIXS (resonant inelastic x-ray scattering) _operando_ studies based on a 3-electrode electrochemistry flow-cell located at the HZB's branch of the EMIL beamline. The modular design of the measurement chamber ensures an easy connection to the newly developed high-transmission soft x-ray emission spectrometer, which covers the photon range between the Li and the Si K-edge with an energy resolving power > 1500.

We present the current status of the instrumentation in EMIL and the first spectroscopic results on our path towards the _in-situ_ characterization of real-world electrocatalysts under operating conditions.

CPP 49.10   Wed 17:30   C 230

Lab-on-a-chip - Electron paramagnetic resonance goes mini — •Silvio Künstner[1], Benedikt Schlecker[2], Anh Chu[3], Jannik Möser[1], Alexander Schnegg[1], Jens Anders[3], and Klaus Lips[1] — [1]Berlin Joint EPR Laboratory, Helmholtz-Zentrum Berlin für Materialien und Energie — [2]Insitut für Mikroelektronik, Universität Ulm — [3]Institut für Theorie der Elektrotechnik, Universität Stuttgart

Electron paramagnetic resonance (EPR) spectroscopy is one of the most powerful techniques used in physics, chemistry and materials science. It is a non-invasive technique sensitive to paramagnetic species, especially suited for the study of paramagnetic states in thin-film materials and at surfaces involved in chemical reactions. However, the current design of EPR spectrometers using a microwave (mw) resonator limits the versatility for operando measurements. Here, we present a radical new design of a miniaturised EPR spectrometer, implemented on a microchip (EPR-on-a-chip). EPRoC is no longer restricted by the boundary condition of a resonator and has a three order of magnitude higher spin sensitivity than conventional EPR. The sensor is a single coil scaled-down in size to a few 10-100 micrometer. Different from conventional EPR, EPRoC is carried out by sweeping the mw frequency instead of the magnetic field, enabling operation with a permanent magnet. Due to its compactness, EPRoC can be incorporated in conventional growth reactors, (electro)chemical cells or in harsh UHV environments. We will review the recent success of operando EPRoC, discuss the detection principle and demonstrate its superior sensitivity, and present first time-domain experiments with nanosecond resolution.