CPP 45: Emerging Topics in Chemical and Polymer Physics, New Instruments and Methods

Time: Thursday 9:30–12:00 Location: ZEU 255

CPP 45.1 Thu 9:30 ZEU 255

Structural effects of many-body van der Waals interactions: from small molecules to polymers — ●Raul Ian Sosa, Mario Galante, and Alexandre Tkatchenko — Department of Physics and Materials Science, University of Luxembourg, L-1511 Luxembourg City, Luxembourg.

Van der Waals (vdW) dispersion interactions are among the key determinants of structure, stability, and dynamics in a wide range of systems such as supramolecules, complex interfaces and low-dimensional nanostructures. Most commonly used approaches for incorporating vdW interactions in extended systems rely on pairwise approximations, neglecting the quantum many-body nature of molecular interactions [Quantum Chem. 114, 1157 (2014), Science. 350, 6257 (2015)]. Recent simulations which incorporate Many-Body Dispersion (MBD) show that a many-body description of van der Waals can be essential for properly describing extended molecules and molecular solids [Physical Review Letters. 108, 23 (2012)]. These effects often manifest themselves as increased ranges of interaction and collective dynamics in a way that is highly dependent on the molecular structure [Nat Comm. 11, 1651 (2020)]. Here we will present a systematic analysis on the impact that many-body interactions have on the range of van der Waals forces in polymers, one dimensional nanostructures and small molecules, making a particular emphasis on the structural dependence of such interactions.

CPP 45.2 Thu 9:45 ZEU 255

Bulk magneto-mechanical response of magnetic filaments in applied magnetic fields — ●Deniz Mostarac and Sofia Kantorovich — Univesity of Vienna,Vienna,Austria

Incorporating magnetic nanoparticles within permanently cross-linked structures, opens up the possibility for synthesis of complex, highly magneto-responsive systems. Magnetic filaments (MFs) are polymer-like chains of magnetic, nano-sized colloids, and are a promising platform for engineering novel, magnetically controlled filtering and flow control elements in micro-fluidic devices. The interplay between central attraction forces, and anisotropic, dipolar interactions is the key factor when tuning the macroscopic response of MFs. In this contribution, we show that taking in to account nonlinear contributions to the magnetisation of super-paramagnetic colloids is essential to capture the bulk, magneto-mechanical response of MFs consisting of magnetisable colloids. In the presence of central attraction forces, we report substantial differences in the equilibrium structure of filament clusters, and the mechanical and magnetic response of filament suspensions, to static and time dependant magnetic fields. While on the level of a single filament, magneto-mechanical properties are in general more dependant on the crosslinking approach than on the magnetic nature of colloids, in bulk, the long-range nature of dipolar interactions in quasi-infinite systems makes up for a tremendous difference, and underlines the necessity of a sophisticated model of magnetic colloids that can be magnetised by the presence of magnetic and dipole fields.

CPP 45.3 Thu 10:00 ZEU 255

Improving the Electrochemically Assisted Surfactant Assembly of Vertically Aligned Mesoporous Silica Films — ●Gilles Möhl[1,2], Samuel Fitch[2], Li Shao[2], Jonathan Rawle[3], Ralph Gilles[1], Guy Denuault[2], Tauqir Nasir[2], Yisong Han[4], Richard Beanland[4], Ruomeng Huang[2], Yasir Noori[2], Philip Bartlett[2], and Andrew Hector[2] — [1]FRM II/TUM, München, Deutschland — [2]UOS, Southampton, United Kingdom — [3]Diamond Light Source, Harwell, United Kingdom — [4]Warwick University, Coventry, United Kingdom

Mesoporous silica films are typically produced by evaporation-induced self-assembly (EISA), but vertical alignment of the pores to the substrate remains difficult. Hexagonal arrays of vertically aligned mesopores can be made by electrochemically assisted surfactant assembly (EASA). The self-assembly of a cationic surfactant (cetyltrimethylammonium bromide), which relies on the production of hydroxide species close to the substrate, also leads to the formation of spheroidal surface aggregates, limiting the obtainable film thickness to a few hundreds of nm. In this work, we show the results obtained from operando Grazing incidence small angle X-ray scattering (GISAXS) experiments done during the EASA of silica, following the evolution of the structure for-

mation in real time with sub-second time resolution. We include the findings from operando pH measurements using an electrochemical microsensor, revealing the temporal evolution of the hydroxide concentration close to the electrode. This enabled us to adapt the EASA protocol to reduce aggregate formation and increase film thickness.

CPP 45.4 Thu 10:15 ZEU 255

Pair-wise dependences of morphological descriptors provide fingerprint of pore structure in cellulose-based materials — ●Karin Zojer[1], Matthias Neumann[2], Phillip Gräfensteiner[2], Eduardo Machado Charry[1], Andre Hilger[3], Ingo Manke[3], Ulrich Hirn[4], and Volker Schmidt[2] — [1]Institute of Solid State Physics,Graz University of Technology, Petersgasse 16, 8010 Graz, Austria — [2]Institute of Stochastics, Ulm University, Helmholtzstrasse 18, 89069 Ulm, Germany — [3]Institute of Applied Materials, Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany — [4]Institute of Bioproducts and Paper Technology, Graz University of Technology, Inffeldgasse 23, 8010 Graz, Austria

Predicting the macroscopic behavior of heterogeneous porous materials from their microscopic structure is formidably challenging, because local morphological microstructure descriptors markedly and erratically vary across a sample. We demonstrate how to quantify cross relationships between pore space-related descriptors and to use these relations to distinguish complex pore spaces at a glance, alike using fingerprints. The key idea is to map the actual microstructure on a multivariate probability distribution. The latter contains strongly compressed information to reproduce the spatial variations of selected properties and their pair-wise interdependencies. Using R-vine copulas, we will exemplarily construct such a distribution of the local morphological descriptors porosity, thickness, surface area per volume and pathway tortuosity for the measured microstructure of paper sheets.

CPP 45.5 Thu 10:30 ZEU 255

***Ab initio* cavity QED - modifying chemistry with strong light-matter interaction** — ●Christian Schäfer[1,2], Enrico Ronca[3], Johannes Flick[4,5], Prineha Narang[5], and Angel Rubio[2,4] — [1]Department of Microtechnology and Nanoscience, MC2, Chalmers University of Technology, 412 96 Göteborg, Sweden — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — [3]Istituto per i Processi Chimico Fisici del CNR (IPCF-CNR), Via G. Moruzzi, 1, 56124, Pisa, Italy — [4]Center for Computational Quantum Physics (CCQ), The Flatiron Institute, 162 Fifth Avenue, New York NY 10010, USA — [5]John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA

The alchemical dream of altering a given material on demand into something desirable is at the very heart of chemistry. Optical-Cavity environments provide a novel handle to non-intrusively control materials and chemistry. The self-consistent interaction between complex electromagnetic environments and realistic materials gave birth to a new discipline, sometimes referred to as 'ab initio QED', on the interface of condensed matter, chemistry and quantum optics.

I will provide a brief introduction into this newly emerged field and illustrate how chemical reactions can be controlled [1] with optical cavities.

[1] Schäfer, C., Flick, J., Ronca, E., Narang, P., and Rubio, A., arXiv:2104.12429 (2021).

CPP 45.6 Thu 10:45 ZEU 255

Structural Descriptors for Constructing High-Dimensional Neural Network Potentials — ●Moritz R. Schäfer[1,2], Jonas A. Finkler[3], Stefan Goedecker[3], and Jörg Behler[1,2] — [1]Lehrstuhl für Theoretische Chemie II, Ruhr-Universität Bochum, 44780Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research AllianceRuhr, 44780 Bochum, Germany — [3]Basel University, Department of Physics, Klingelbergstrasse 82, 4056 Basel, Switzerland

High-dimensional neural network potentials (HDNNPs) are a well established method to efficiently compute close-to ab initio-quality energies and forces for performing large-scale molecular dynamics simulations of complex systems. In this method, the total energy is constructed as a sum of environment-dependent atomic energy contribu-

tions. Also electrostatic interactions based on flexible atomic charges can be included. Both components crucially depend on the quality of the structural descriptors employed to characterize the local atomic environments. Here we investigate the combination of atom-centered symmetry functions with the recently proposed overlap matrix descriptor. Moreover, the advantages and disadvantages of both descriptors are discussed and illustrated for benchmark systems.

CPP 45.7   Thu 11:00   ZEU 255

Solving inverse transport problems across irregular interfaces of sorptive porous media via physics-informed neural networks — •Alexandra Serebrennikova and Karin Zojer — Institute of Solid State Physics, TU Graz, Petersgasse 16, 8010, Graz, Austria

We show how state of the art extended physics-informed neural networks serve us to solve inverse transport problems with jump conditions across irregular interfaces. This approach reveals the material constants which govern reactive diffusion of organic volatiles migrating across an interface between porous sorptive packaging and food media if we provide experimental data and a transport model.

In such scenarios, associated differential equations (PDE) imply jumps not only in the solution, but also in the solution gradient across interfaces. The idea is to use multiple NN to construct the solution; each NN approximates the solution function of the PDE associated to a domain within the defined interfaces. The networks are coupled across interfaces such that the boundary conditions are satisfied.

As NNs are required to fit underlying physics by minimizing PDE residuals, they are inherently suited to solve inverse problems for the parameters involved in the equations. As further benefit, the discretized experimental data can be represented with a continuous function which offers a meshfree and compact surrogate model for the solution function.

15 min. break

CPP 45.8   Thu 11:30   ZEU 255

Aging-driven compositional changes in Li-ion batteries — •Dominik Petz[1,2], Peter Müller-Buschbaum[1,2], and Anatoliy Senyshyn[1] — [1]MLZ, TUM, Garching — [2]TUM School of Natural Sciences, Chair of Functional Materials, Garching

Electrochemical cycling of lithium-ion batteries is supplemented by the active transport of lithium ions and electrons, which are exchanged between the cathode and anode material. Besides material properties, such exchange is facilitated by the parameters of electrochemical cell like electrode dimensions and geometry, current density, temperature, etc. Such parameters are neither uniformly distributed nor static in general and, therefore, serve as a factor stabilizing heterogeneous states in Li-ion batteries typically reflected in the lithium concentration profiles in the electrodes. Thus, cell aging directly affects the distribution of the lithium-ions in the graphite anode of 18650-type lithium-ion batteries. Lithium heterogeneities (often reflected in loss of lithium inventory) are directly related to the electrolyte filling level. In order to quantify and correlate them simultaneously, a set of cells at different state-of-health was studied non-destructively using spatially resolved neutron powder diffraction. Experimental results unambiguously revealed changes of the lithium and electrolyte distribution versus cell state-of-health in a series of commercial 18650-type lithium-ion battery.

CPP 45.9   Thu 11:45   ZEU 255

Dedoping of PEDOT:PSS using amines to shift the threshold voltage in OECTs — •Laura Teuerle, Rakesh Nair, Hans Kleemann, and Karl Leo — IAPP Dresden

With an increasing importance of organic semiconductors, due to their flexibility, transparency and possible biocompatibility, more applications and device classes emerge.

One of them - the organic electochemical transistor (OECT) can be utilized in neuromorphic computing and printed digital logic. The most commonly used -PEDOT:PSS is a p-type semiconductor, which leads to normally-on type transistors. However, normally-off type devices are needed for a successful application in logic circuits.

We show that a chemical dedoping method involving amines leads to a shift in threshold voltage to zero and even negative in the corresponding devices.

Different processing methods for the dedoping process and device building are studied, which involve photolithography, inkjet printing and spraycoating.

Furthermore, IV measurements for device characterization were taken.

The results show that the studied chemical dedoping of -PEDOT:PSS can be a viable method to create normally-off type transistors for an application in logic circuits.