BP 18: Biomaterials and Biopolymers (joint session BP/CPP)

Time: Wednesday 9:30–13:00 Location: H 1028

Invited Talk BP 18.1 Wed 9:30 H 1028

Production and applications of artificial spider silk fibers and hydrogels — •Anna Rising — Department of Anatomy Physiology and Biochemistry Swedish University of Agricultural Sciences Uppsala 75007, Sweden — Department of Biosciences and Nutrition, Karolinska Institutet, Neo, Huddinge 14183, Sweden

Spider silk, nature's high-performance fiber, represents an attractive material for many different applications. However, production of the spider silk proteins (spidroins) is problematic due to their repetitiveness and propensity to aggregate.

We have developed a E.coli based production method that generates unprecedented amounts of correctly folded and soluble spidroins. A biomimetic spinning method combined with a protein engineering strategy, result in artificial spider silk fibers that match the toughness of native spider silk. The fibers have successfully been used to guide the extension of neurites in cell culture assays.

In addition, we have discovered that the recombinant spidroins rapidly form self-supporting and transparent hydrogels when incubated at 37 °C. The gelation is associated with the formation of nano-sized fibrils, and spidroin fusion proteins form hydrogels with intact functions of the fusion moieties. By varying the protein concentration, the compressive modulus of the hydrogels can be tuned to match that of skeletal muscle, myocardium and cartilage, respectively. In addition, human mesenchymal stem cells are viable after being encapsulated in the gels, and continuous release of biologics can be achieved as exemplified by an encapsulated cell line producing progranulin.

BP 18.2 Wed 10:00 H 1028

Understanding the molecular determinants of chitin-protein interactions in the arthropod cuticle - a single-molecule approach — •Ayesha Talib[1,2], Yael Politi[2], and Kerstin G. Blank[1,3] — [1]Max Planck Institute of Colloids and Interfaces, Mechano(bio)chemistry, Am Mühlenberg 1, 14476 Potsdam, Germany — [2]Technische Universität Dresden, CMCB, B CUBE, Tatzberg 41, 01307 Dresden, Germany — [3]Johannes Kepler Universität, Institute of Experimental Physics, Altenberger Straße 69, 4040 Linz, Austria

In the cuticle of arthropods, structural proteins and chitin fibers form a composite material with anisotropic mechanical properties. The molecular parameters that define the chitin-protein interaction are largely unknown. To answer the fundamental question of what controls cuticle mechanical properties, a molecular strategy is employed that integrates protein engineering with single-molecule force spectroscopy. Chitin binding domains (CBDs) from the spider Cupiennius salei have been identified and expressed recombinantly to compare the strength of the protein-chitin interaction. For CBD present in all spider tissues, we investigated three overlapping consensus motifs RR-1, RR-2 and CB-4. Pull-down assays and single-molecule force spectroscopy suggest that the RR-1 motif does not bind to chitin, whereas similar binding strength is observed for the RR-2 and CB-4. We observe a fast dissociation rate, suggesting that CBDs facilitate energy dissipation upon deformation. Our ultimate goal is to correlate molecular properties with the mechanical function of the composite and to synthesize artificial analogues with tunable mechanical properties.

BP 18.3 Wed 10:15 H 1028

Characterizing bursting spider silk coacervates with micropipette aspiration — •Isabell Tunn[1], Grégory Beaune[2], Jennifer Tersteegen[1], Jaakko V.I. Timonen[2], Francoise Brochard-Wyart[3], and Markus B. Linder[1] — [1]Department of Bioproducts and Biosystems, Aalto University, Finland — [2]Department of Applied Physics, Aalto University, Finland — [3]Institute Curie, Université Paris Sciences et Lettres, Sorbonne Université, Laboratoire Physico Chimie Curie, France

Hollow or core-shell coacervates composed of biomolecules have been reported to serve essential intracellular functions. Recently, numerous hollow and core-shell coacervates have been bioengineered in vitro opening new avenues for their application as drug delivery systems or vessels for chemical reactions. However, the relationship between the molecular structure and the biophysical properties of these coacervates remains largely unexplored. Thus, we characterized the biophysical properties of a set of five bioengineered spider silk protein coacervates using micropipette aspiration. Upon aspiration coacervates can burst like vesicles, demonstrating that protein forms a dense layer (shell) on the surface of the coacervate. To analyse the aspiration and bursting of the hollow coacervates we developed a model, which allows to calculate the surface and bulk viscosity and to estimate the thickness and viscosity of the shell. We anticipate that our model will aid in understanding the formation and properties of hollow coacervates and will facilitate their use as drug delivery systems, reaction vessels as well as material building blocks.

BP 18.4 Wed 10:30 H 1028

Connecting protein architecture to their emergent droplet properties — •Zhouyi He, Jens-Uwe Sommer, and Tyler Harmon — Leibniz-Institut für Polymerforschung, Institut Theorie der Polymer, 01069, Dresden, Germany

Spatial organization is a fundamental characteristic of biological systems. Biomolecular condensates (droplets) is one class of spatial organization. Understanding how these droplets arise from molecular interactions remains a complex challenge. To address this, we focused on a specific model system consisting of two multivalent proteins with folded domains and disordered linkers which co-phase separate into droplets. We employed coarse-grained simulations to investigate how structural modularity impacts the phase diagram and material properties of these droplets. Our study focuses on the material properties: density, viscosity, and network architecture. The introduction of a coiled-coil domain substantially alters the phase behavior and material properties of the resulting droplets. The stiffness of this domain plays an important role in preventing self-loop closure and thus promoting phase separation, which enables a certain degree of orthogonal design across the various material properties we explored. Our research yields insights into the phase behavior of biomolecular condensates and provides guidelines for the modulation of droplet material properties.

BP 18.5 Wed 10:45 H 1028

Strain-Controlled Critical Slowing Down in the Rheology of Disordered Networks — •Abhinav Sharma[1,2], Jordan Shivers[3], and Fred MacKintosh[3] — [1]Mathematisch-Naturwissenschaftlich-Technische Fakultät, Universität Augsburg, 86159 Augsburg — [2]Leibniz Institute für Polymerforschung, Dresden — [3]Department of Chemical and Biomolecular Engineering, Rice University, Houston, Texas 77005, USA

Networks and dense suspensions frequently reside near a boundary between soft (or fluidlike) and rigid (or solidlike) regimes. Transitions between these regimes can be driven by changes in structure, density, or applied stress or strain. In general, near the onset or loss of rigidity in these systems, dissipation-limiting heterogeneous nonaffine rearrangements dominate the macroscopic viscoelastic response, giving rise to diverging relaxation times and power-law rheology. Here, we describe a simple quantitative relationship between nonaffinity and the excess viscosity. We test this nonaffinity-viscosity relationship computationally and demonstrate its rheological consequences in simulations of strained filament networks and dense suspensions. We also predict critical signatures in the rheology of semiflexible and stiff biopolymer networks near the strain stiffening transition.

BP 18.6 Wed 11:00 H 1028

Mechanically biomimetic hydrogel suggests fundamental viscoelastic constraints in intracellular mechanics — •Dorian Marx, Bart Eduard Vos, Till Moritz Münker, and Timo Betz — Drittes Physikalisches Institut - Biophysik, Georg-August-Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany

The currently best fitting model for the viscoelastic mechanical properties of cellular cytoplasm lacks a connection to physical parameters. To establish this connection, we find and utilize a viscoelastic polyacrylamide-based hydrogel with cytoplasm-like mechanical properties. The variation of experimental parameters reveals the connection of their physical interpretations to the mechanical outcome of the measurement. The used viscoelastic hydrogel enables the comparison of different measurement methods spanning length scales of micrometers (optical tweezers) to centimeters (rheometer) and can serve as a calibration material that is neither ideally elastic nor ideally viscous. Furthermore, a detailed analysis of cellular and hydrogel data uncovers

striking correlations of model parameters that closely match between the chemically and physically distinct materials. The results motivate further investigations into the theoretical reasons for the correlations and applications in building artifical cells.

15 min. break

BP 18.7 Wed 11:30 H 1028

Dynamic DNA origami nanopores — •Anna Baptist[1], Ze Yu[2], Sabina Caneva[2], and Amelie Heuer-Jungemann[1] — [1]MPI of Biochemistry, Martinsried, Germany — [2]TU Delft, The Netherlands

Nanopores are nanoscale structures that form channels across membranes and enable the translocation of molecules. Inspired by naturally occurring pore-forming proteins, different types of artificial nanopores have been created. The DNA origami technique allows for the fabrication of DNA nanostructures with precise control over shape and size that can be modified with a variety of functional molecules. Thus, DNA origami provides a platform for the customized design of nanoscale pores with different channel diameters that can be equipped with anchoring molecules for insertion into lipid membranes. Such nanopores have potential applications in single-molecule sensing, sorting of molecules depending on their sizes or for the fabrication of artificial cells. However, most nanopores created so far are static with a fixed pore diameter. Here, we present a large dynamic DNA origami nanopore that can be mechanically and reversibly switched between different conformations via strand displacement, offering three different pore sizes. After their successful insertion into the lipid bilayer, these nanopores form transmembrane channels with varying diameters depending on their conformation and can be used to control the transport of differently sized molecules across the lipid membrane. Such stimuli-responsive, actuatable nanopores are excellent mimics of complex natural occurring pores, while enabling a higher level of control and a more modular and easily adaptable design.

BP 18.8 Wed 11:45 H 1028

Nanotextured Surfaces Based on DNA — •Irina Martynenko and Tim Liedl — Faculty of Physics, Ludwig-Maximilians-University, 80539 Munich, Germany

A longstanding goal of material scientists is to fabricate functional materials in which nanoscale objects are precisely positioned on macroscale surfaces. This can be achieved by a combination of bottom-up techniques, such as molecular self-assembly of DNA origami, and top-down lithographic methods. Through DNA origami placement (DOP) on lithographically patterned surfaces a variety of nanoscale components such as organic dyes, proteins or nanoparticles, have already been patterned on large-scale arrays [1, 2]. However, any DOP methods developed so far were limited to two-dimensional DNA origami structures and thus resulted in flat patterns and arrays only. Here we extend DOP to the third dimension through positioning of three-dimensional DNA origami onto nanometer-precise patterns over micro- and even millimeter scales [3]. We demonstrate that our method can produce surfaces nanotextured with three-dimensional hybrid DNA-silica structures with controllable heights up to 50 nm and a feature size down to ~ 6 nm. We believe that the presented strategy can be used for the assembly of a wide range of materials from metals and semiconductors to functional biomolecules arranged in virtually any three-dimensional geometry on large-scale substrates. [1] R. Kershner, Nat Nanotechnol (2009) [2] A. Gopinath, et al., Nature (2016) [3] I. Martynenko et al., Nat Nanotechnol (2023)

BP 18.9 Wed 12:00 H 1028

DNA-Origami Diamond Crystal with photonic bandgap in the UV Range — •Xin Yin[1], Gregor Posnjak[1], Paul Butler[2], Oliver Bienek[2], Mihir Dass[1], Ian Sharp[2], and Tim Liedl[1] — [1]Ludwig-Maximilian-Universität München, Germany — [2]Walter Schottky Institute, Technical University Munich, Germany

Diamond lattice photonic crystals possess a broad complete photonic bandgap, although its manufacturing has proven challenging. [1] We showcase a DNA origami diamond crystal with 170 nm periodicity. [2] DNA origami is a technique that allows the rational design of complex geometries on the nanoscale, [3] which we apply to build tetrapod single units for the crystal. Pristine crystal formation requires careful control of interactions between the monomers. The thus-formed crystal undergoes silicification, via a wet chemistry method, for enhanced mechanical stability, followed by TiO2 coating via atomic layer deposition (ALD). The latter process is required to increase the refractive index and thus open the photonic bandgap. Optical measurement reveals a reflection band in UV range, with the peak red shifting as the coating thickness increases. These results align well with simulations predicting the structure's photonic properties. [1] R. K. Cersonsky, J. Antonaglia, B. D. Dice, & S. C. Glotzer. Nature communications, 12(1), 2543. [2] G. Posnjak, X. Yin, P. Butler, O. Bienek, M. Dass, I. D. Sharp, & T. Liedl, arXiv preprint arXiv:2310.10884. [3] P. W. Rothemund. Nature, 440(7082), 297-302.

BP 18.10 Wed 12:15 H 1028

Scaling properties of RNA as a branched polymer — Domen Vaupotič[1], Angelo Rosa[2], Luca Tubiana[3], and •Anže Božič[1] — [1]Jožef Stefan Institute, Ljubljana, Slovenia — [2]SISSA, Trieste, Italy — [3]University of Trento, Trento, Italy

Formation of base pairs between the nucleotides of an RNA sequence gives rise to a complex and often highly branched RNA structure. While numerous studies have demonstrated the functional importance of the high degree of RNA branching, its topology remains largely unexplored. We use the theory of randomly branching polymers to explore the scaling properties of RNAs by mapping their secondary structures onto tree graphs. Focusing on random RNA sequences of varying lengths, we determine the two scaling exponents related to their topology of branching. Our results indicate that ensembles of RNA secondary structures are characterized by annealed random branching and scale similarly to self-avoiding trees in three dimensions. We further show that the obtained scaling exponents are robust upon changes in nucleotide composition, tree topology, and folding energy parameters. Finally, we demonstrate how the scaling exponents can be obtained from the distributions of the related topological quantities of individual RNA molecules with fixed length. In this way, we establish a framework to study the branching properties of RNA and compare them to other known classes of branched polymers. By understanding the scaling properties of RNA structure we aim to improve our understanding of the underlying principles and open up the possibility to design RNA sequences with desired topological properties.

BP 18.11 Wed 12:30 H 1028

The role of receptor uniformity in multivalent binding — •Xiuyang Xia[1,2], Ge Zhang[3], Massimo Pica Ciamarra[1], Yang Jiao[4], and Ran Ni[1] — [1]Nanyang Technological University, Singapore — [2]Ludwig-Maximilians-Universität München, Munich, Germany — [3]City University of Hong Kong, Hong Kong, China — [4]Arizona State University, Tempe, USA

Multivalency is prevalent in various biological systems and applications due to the superselectivity that arises from the cooperativity of multivalent binding. Traditionally, it was thought that weaker individual binding would improve the selectivity in multivalent targeting. Here using analytical mean field theory and Monte Carlo simulations, we discover that for receptors that are highly uniformly distributed, the highest selectivity occurs at an intermediate binding energy and can be significantly greater than the weak binding limit. This is caused by an exponential relationship between the bound fraction and receptor concentration, which is influenced by both the strength and combinatorial entropy of binding. Our findings not only provide new guidelines for the rational design of biosensors using multivalent nano-particles but also introduce a new perspective in understanding biological processes involving multivalency.

BP 18.12 Wed 12:45 H 1028

Salt Effects on Caffeine Across Concentration Regimes — Stefan Hervo-Hansen[1,4], Jakub Polák[2], Markéta Tomandlová[2], Joachim Dzubiella[3], •Jan Heyda[2], and Mikael Lund[4] — [1]Chem. Engineering Div., Osaka Uni., Toyonaka, Osaka 560-8531, Japan — [2]Physical Chem. Dpt., UCT Prague, Technická 5, CZ-16628 Praha 6, Czechia — [3]Physikalisches Inst., Albert-Ludwigs Uni. Freiburg, Hermann-Herder-Straße 3, D-79104 Freiburg im Breisgau, Germany — [4]Theoretical Chem. Div., Lund Uni., Lund SE 221 00, Sweden

This theoretical contribution is motivated by the early work of Charles Tanford, which led to the discovery that molecular surface motifs solvation is proportional to the solvent accessible surface area (SASA). Importantly, later studies have shown that the proportionality constant varies with salt *concentration* and *type*. Using multi-scale computer simulations combined with vapor-pressure osmometry on caffeine-salt solutions, we reveal that this SASA description captures a rich set of molecular driving forces in ternary solutions at changing *solute* and *osmolyte* concentrations.

Central to this theoretical work is a new potential energy function

that depends on the instantaneous surface area, salt type, and concentration. Used in *e.g.* Monte Carlo simulations, this allows for a highly efficient exploration of many-body interactions and the resulting thermodynamics at elevated solute and salt concentrations.

This protocol opens the path to account for salt effects on the solvation thermodynamics of molecules of all sizes, including the *salting-in* or *salting-out* of aqueous bio(macro)molecules.