CPP 72: Polymer Networks and Elastomers

Time: Wednesday 15:00–17:30 Location: ZEU 114

Invited Talk CPP 72.1 Wed 15:00 ZEU 114

Physics of Fiberboids — •Igor Kulic — CNRS, Institut Charles Sadron, 23 rue du Loess BP 84047, 67034 STRASBOURG Cedex 2

Fiberboids are filaments trapped at the interface between two phases, able of harnessing energy and matter fluxes across the interface to produce a rolling-like self-propulsion. We experimentally demonstrate several common driving mechanisms and develop the physical framework for understanding the complex active dynamics of fiberboids. The phenomenon is highly generic and plays a role across scales, from the macroscopic realm down to the nanoscale, where it gives rise to chemical fuel-driven surface motility of various filamentous viruses.

[1] A. Baumann, A. Sánchez-Ferrer, L. Jacomine, P. Martinoty, V. Le Houerou, F. Ziebert & I. M. Kulić , Motorizing fibres with geometric zero-energy modes , Nature Materials, 17, 523 (2018) [2] F. Ziebert & I. M. Kulic, Frustriert in Bewegung, Physik Journal 11 (2018)

CPP 72.2 Wed 15:30 ZEU 114

Swelling of tendomer networks: a model system for topological gels — •Toni Müller[1,2], Michael Lang[1], and Jens-Uwe Sommer[1,2] — [1]Leibniz Institute of Polymer Research Dresden, Germany — [2]TU Dresden, Germany

A polyrotaxane (PR) consists of many small cyclic molecules (”rings”) threaded onto a long polymer chain where stopper monomers at the chain ends prevent detachment of the rings. Polymer networks can be constructed by linking the rings in solution of PRs. We discuss the tendomer as a model system where only their very first rings of the PRs are connected while all remaining rings, m, form a one-dimensional repulsive gas along the PR. Pulling on pairs of chain ends next to the connected rings induces a compression of the sliding rings confined on the two chains. We investigate the force extension relation of the tendomers by computer simulations and compute numerically the exact solution of full partition function in the constant force ensemble. Our results show that tendomers are stiff for low applied forces and soften dramatically when reaching a stall force that is triggered by the density of non-crosslinked rings on the PRs. For large deformations, the finite extensibility of the chains results again in a lower susceptibiliy regarding an applied force [1]. These results are inserted into the Flory-Rehner model predicting the equilibrium degree of swelling, Q, of gels made by crosslinking tendomers. We find that Q scales with m by a power law of 3/4 and that entanglements are negligible.

[1] Müller, T., Sommer, J.-U., Lang, M. (2019) Soft Matter, 15(18), 3671-3679

CPP 72.3 Wed 15:45 ZEU 114

An Analysis of the Gel Point of Polymer Model Networks by Computer Simulations — •Michael Lang and Toni Müller — Leibniz Institut für Polymerforschung Dresden, Hohe Straße 6, 01069 Dresden

The gel point of end-linked model networks is determined from computer simulation data. It is shown that the difference between the true gel point conversion, $p_{\rm c}$, and the ideal mean field prediction for the gel point, $p_{\rm c,id}$, is a function of the average number of cross-links per pervaded volume of a network strand, P, and thus, contains an explicit dependence on junction functionality f for stoichiometric model networks. On the contrary, the amount of intramolecular reactions at the gel point is independent of f in first approximation and exhibits a different power-law dependence on the overlap number of elastic strands as compared to $p_{\rm c} - p_{\rm c,id}$. Therefore, the delay of the gel point, $p_{\rm c} - p_{\rm c,id}$, cannot be predicted from intramolecular reactions and vice versa in contrast to a long standing proposal in the literature about polymer networks.

CPP 72.4 Wed 16:00 ZEU 114

Understanding the static and dynamic behaviour of stars forming reversible networks — •Kiran Suresh Kumar[1,2], Toni Müller[1,2], Jens-Uwe Sommer[1,2], and Michael Lang[1] — [1]Leibniz-Institut für Polymerforschung Dresden, Institut Theorie der Polymere, Hohe Strasse 6, 01069 Dresden, Germany — [2]Institute für Theoretische Physik, Technische Universität Dresden, Zellescher Weg 17, 01069 Dresden, Germany

Reversible networks break and reform continously allowing the material to flow and self-heal on long time scales while being a solid on short times. Recent experiments and simulation studies find an apparent anomalous superdiffusive regime in reversible networks by analyzing Forced Rayleigh Scattering (FRS) data [1-3]. The molecular origin of this superdiffusive regime is not yet fully understood. In our contribution, we approach this problem by computer simulations of FRS experiments in reversible networks using the Bond Fluctuation Model. We analyze the static properties and the connectivity of individual stars and develop a thermodynamic model for the statistics of connections. We analyze dynamic properties of individual stars and compute the collective relaxation as accessible in FRS. Our goal is to develop a model based upon the molecular statistics that allows to quantitatively predict the collective dynamics of the reversible network.

[1] Tang, S.; Wang, M.; Olsen , B. D. J. Am. Chem. Soc. 2015, 137, 3946-3957. [2] Tang, S.; Habicht, A.; Li, S.; Seiffert, S.; Olsen, B. D. Macromolecules 2016, 49, 5599-5608. [3] Ramirez, J.; Dursch, T. J.; Olsen , B. D. Macromolecules 2018, 51, 2517-2525.

CPP 72.5 Wed 16:15 ZEU 114

Constitutive modelling of magneto-sensitive elastomers — •Sanket Chougale, Dirk Romeis, and Marina Saphiannikova — Institute Theory of Polymers, IPF, Dresden, Germany

One type of field-controllable materials are magneto-sensitive elastomers (MSEs) with magnetically switchable properties. They consist of magnetizable particles incorporated within an elastomer matrix. In the presence of an external magnetic field, the induced magnetic interactions and the corresponding particle rearrangements change the mechanical properties substantially. This leads to change in the macroscopic shape of MSE. The magnetic field also introduces a mechanical anisotropy with an axis of symmetry along the field. As a results, MSE can be studied phenomenologically as a transversely isotropic material. Our aim to derive an effective material model from the free energy of MSE in the dipole approximation. As a first step we consider uniaxial deformations parallel and perpendicular to the applied field direction. We compare the stresses derived from the physical model of MSE with the stresses derived from the phenomenological model and find a good agreement. This allows us to extract the coefficients of transversely isotropic material model. We thank the DFG for financial support through RTG-2430.

CPP 72.6 Wed 16:30 ZEU 114

Nonlinear mechanics of physically crosslinked elastomers: from molecular simulations to network models — •Joerg Rottler[1] and Amanda Parker[2] — [1]Department of Physics and Astronomy, University of British Columbia, Vancouver, Canada V6T 1Z1 — [2]Data 61, CSIRO, Melbourne, Australia

Thermoreversible (physical) crosslinking is a versatile strategy for improving the strength and toughness of elastomers that also permits self-healing upon thermal cycling. There is no universally applicable theory for predicting their mechanical response because the mechanical response is governed by the microscopic polymer chain conformations during deformation that cannot easily be measured experimentally. Here, we present a family of entropic network models that account for the stress contributions arising from chain crosslinks as well as entanglements by coupling analytical expressions for the strain energy density directly with chain deformations obtained through molecular dynamics simulations. Our theory quantitatively reproduces the macroscopic stress response of simulated linear and star block copolymer elastomers well into the nonlinear regime. The simulations reveal the evolution of entanglements and how the breakup of physical crosslinks contributes to additional strain hardening. Although developed for a classic sphere forming ABA block copolymer system, our approach can readily be applied to any (bio)macromolecular network in which a rigid phase anchors polymer chains otherwise forming a rubbery matrix.

15 min. break

CPP 72.7 Wed 17:00 ZEU 114

Morphology of adhesive creases — •Michiel van Limbeek[1], Martin Essink[2], Anupam Pandey[3], Jacco Snoeijer[2], and Stefan Karpitschka[1] — [1]Max Planck Institue for Dynamics and Selforgization, Göttingen, Germany — [2]University of Twente, Enschede,

the Netherlands — [3]Cornell University, Ithaca, the Unitied States of America

The compression of an elastic material beyond a certain strain turns the free surface to become unstable. The material makes a sharp fold of the surface onto itself, releasing elastic energy in the bulk. The resulting morphologies are observed in growing tissues and swelling gels. Self adhesion within the folded region is known to affect nucleation and hysteresis: A uncreased sample requires a higher critical strain for creasing than a previously creased one. However, a detailed description of the crease phenomena has remained elusive. Here we resolve the geometry and mechanics of adhesive creases. We combine numerical simulations, analysis and experimental results, where we pay specific attention to the singular edge of the self-contact, which we managed to visualize using confocal microscopy. In the region of self contact, a competition emerges between elastic and surface energies. We compare the morphology for different gel-stiffnesses and it turns out that adhesive creases exhibit a universal shape after proper rescaling. We derive a scaling theory for the aforementioned bifurcation scenario of the hysteresis, explaining the nucleation of adhesive creases.

CPP 72.8 Wed 17:15 ZEU 114

Tough and Instantly Recoverable Hydrogels: Self-Reinforcement by Strain-Induced Crystallization — ●Koichi Mayumi[1], Chang Liu[1], Jiang Lan[1], Takamasa Sakai[2], Hideaki Yokoyama[1], and Kohzo Ito[1] — [1]Graduate School of Frontier Sciences, The University of Tokyo, Chiba, Japan — [2]Graduate School of Engineering, The University of Tokyo, Tokyo, Japan

Most tough hydrogels are reinforced by introducing sacrificial structures that can dissipate input energy [1]. However, since the sacrificial damages cannot recover instantly, the toughness of these gels drops substantially during consecutive cyclic loadings. In this presentation, we will propose a new damageless reinforcement strategy for hydrogels utilizing strain-induced crystallization (SIC). In Slide-Ring (SR) gels where polyethylene glycol (PEG) chains are cross-linked by rings [2,3], crystalline of PEG repetitively forms and destructs with elongation and relaxation, resulting in both excellent toughness of 5.5 to 25.2 MJ/m^3 and 87% to 95% instant recovery of extension energy between two consecutive loading-unloading cycles. The instantly reversible tough hydrogels are promising candidates for applications in artificial connective tissues such as tendon and ligament.

[1] J. P. Gong, Soft Matter, 6, 2583 (2010). [2] K. Ito, Polymer Journal, 39, 489 (2007). [3] L. Jiang, C. Liu, K. Mayumi, K. Kato, H. Yokoyama, K. Ito, Chemistry of Materials, 30, 5013 (2018).