CPP 8 SYMPOSIUM: Polymer networks and beyond: From molecular structure to materials and biological functions III

Zeit: Samstag 14:15–16:40 Raum: TU C243

Hauptvortrag CPP 8.1 Sa 14:15 TU C243

The diffusion of polymers in networks and the importance of network structure — •Mark Geoghegan — Department of Physics and Astronomy, University of Sheffield

The structure of polymer networks has profound implications for their properties. In particular it is not possible to characterise networks by two parameters, as is traditionally done (average number of monomers between crosslinks and volume fraction of network in the preparation state). We consider experimental results (using neutron reflectometry, ion beam profiling, and fluorescence correlation spectroscopy) based at elucidating information on the structure of polymer networks and then describe work investigating the diffusion of homopolymers in dry or solvent swollen polymer networks. Thin film effects will also be discussed, focussing on issues such as the behaviour of sol in networks, as well as the role of interfaces in adhesion.

Hauptvortrag CPP 8.2 Sa 14:45 TU C243

Liquid Crystal Elastomers: Mechanics and Optics — •Heino Finkelmann — Institut für Makromolekulare Chemie, Albert-Ludwigs-Universität Freiburg, 79104 Freiburg

Rubber elasticity is a unique feature of polymer networks or elastomers formed from long polymer chains connected to one another by crosslinkages. An implanting of rod-like mesogenic monomer units into the network chains can induce the liquid crystalline state of the elastomers. The interplay between network chain conformation and liquid crystalline phase creates material with remarkable properties. Detailed investigations proved a direct coupling between chain conformation and anisotropic state of order of the liquid crystalline phase. Depending on the liquid crystalline phase structure eceptional properties are observed: Nematic networks change their macroscopic dimensions when the state of order is modified by external stimuli. This effect can be applied to novel thermo- and opto-mechanical actuators. Chiral nematic (cholesteric) elastomers, because of their birefringence and periodic, helicoidal structure, are 1-d photonic band-gap materials which enable mirrorless lasing. Mechanical deformation of the networks couples to the phase structure and causes a shift of the emission wave length.

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Cholesteric Liquid Crystal Elastomers as Mechanically Tunable Photonic Band Gap Materials — •Jürgen Schmidtke[1], Werner Stille[1], Simon Kniesel[2], and Heino Finkelmann[2] — [1]Physikalisches Institut, Albert-Ludwigs-Univ., Freiburg — [2]Institut für Makromolekulare Chemie, Albert-Ludwigs-Univ., Freiburg

We have studied the effects of uniaxial and biaxial strain on the photonic band structure of an elastomer with cholesteric liquid-crystalline order. Cholesteric liquid crystals (CLCs) act as polarization-sensitive one dimensional photonic crystals. Due to the elastic coupling of the liquid-crystalline phase structure and the polymer network, a CLC elastomer allows for a mechanical tuning of the optical properties.

On application of biaxial strain, the photonic stop band is shifted to shorter wavelengths. The number of director turns along the film normal is found to be a conserved quantity. Using a sample doped with a laser dye, photonic band edge lasing can be mechanically tuned over a wavelength range of about 100 nm.

Application of uniaxial strain perpendicular to the helical axis results in a modification of the photonic band structure, due to a deformation of the cholesteric helix. However, contrary to theoretical predictions, no polarization-independent photonic stop band emerges. Again, the shift of the stop band is affine to the compression along the helical axis. The anisotropic molecular order of the elastomer leads to an anisotropic elastic modulus: Perpendicular to the applied stress, contraction is not isotropic, but to some degree hindered along the direction of the cholesteric helix.

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On the Swelling Behaviour of Polyelectrolyte Networks — •Bernward A. Mann, Christian Holm, and Kurt Kremer — MPI für Polymerforschung, Ackermannweg 10, 55128 Mainz, Germany

Charged Hydrogels have long been essential ingredients in multiple industrial products of our daily life, *e.g.* as superabsorbants. While extensively studied experimentally, theoretical treatment usually applied rather simplistic models to be able to describe the swollen equilibrium state.

Employing molecular dynamics computer simulations, it is now possible to investigate their swelling behaviour independent of analytical assumptions made. In our previous work [1,2], we considered a model network with explicit counterions immersed in good solvent and close to the θ-point, carefully detailing the interplay of the different pressure contributions.

Expanding on this, we are reporting on our findings regarding poor solvent conditions, which drastically alter the swelling behaviour of the polyelectrolyte networks. Not only can the pearl-necklace-structures and the "sausage"regime be found, both known from simulations of single polyelectrolyte chains, but the network connectivity also allows for other structures to be formed, such as *e.g.* candle-like strands.

[1] B. Mann, R. Everaers, C. Holm, K. Kremer, EPL **17**, 786 (2004).
[2] B. Mann, R. Everaers, C. Holm, K. Kremer, *submitted.*

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Dielectric relaxation of gelling polymers — •Alice von der Heydt[1], Henning Löwe[1], Peter Müller[2,1], and Annette Zippelius[1] — [1]Institut für Theoretische Physik, Universität Göttingen — [2]Department of Mathematics, University of California, Irvine

Frequency-dependent dielectric relaxation of gelling polymers was investigated within Rouse dynamics for a macroscopic model network with random polar crosslinks of concentration c drawn from a bond percolation ensemble. Averaged over a crosslink-ensemble at concentration c and uncorrelated dipole orientations, the generic dielectric susceptibility $\chi_c(\omega)$ is given in terms of the ensemble-averaged density of states $D_c(\gamma)$ or the resolvent of the network's connectivity matrix Γ. When the system approaches the sol-gel transition at the critical concentration $c_{\rm crit}$, the growing impact of small relaxation rates is clearly visible in $\chi_c(\omega)$: A crossover in the low-frequency domain from asymptotic Debye behavior at small c to e.g. a cusp for $\mathrm{Re}\,\chi_c(\omega)$ at $\omega = 0$ and criticality can be deduced from the known scaling of $D_c(\gamma)$. For a mean field ensemble, the critical change in the exponents is confirmed by an approach which does not assume scaling. In this case the frequency dependence can be obtained via the resolvent of Γ by numerically solving an exact integral equation which arises in the replica formulation of the disorder average [1].

[1] K. Broderix, T. Aspelmeier, A. K. Hartmann and A. Zippelius, Phys. Rev. E **64** (2001), 021404

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Swelling, Elasticity and Spatial Inhomogeneity of Poly(N-isopropylacrylamide)/Clay Nanocomposite Hydrogels — •Jingjing Nie, Binyang Du, and Wilhelm Oppermann — Institute of Physical Chemistry, Clausthal University of Technology, Arnold-Sommerfeld Str. 4, 38678 Clausthal-Zellerfeld, Germany

Hydrogels were prepared by polymerizing N-isopropylacrylamide without any chemical cross-linker in an aqueous dispersion of exfoliated clay particles. The shear modulus of such gels rises in proportion with clay concentration. This seems to indicate that we have a network where the clay particles act as multifunctional cross-links. The influence of clay concentration, monomer concentration, and preparation temperature on macroscopic and structural properties was systematically investigated by mechanical measurements, swelling measurements and static light scattering. Interestingly, the swelling ratio is fairly independent of clay concentration and also of the other parameters varied. This is in contrast to chemically cross-linked networks and may be traced back to a markedly different microstructure. The static light scattering results reveal two correlation lengths, i.e. two types of spatial inhomogeneities. Their dependence on the parameters used to synthesize the networks will be discussed.

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Application of "Smart" PNIPAM microgels as sensor or actuator — •Thomas Hellweg[1], Frieder Mugele[2] und Regine von Klitzing[3] — [1]TU Berlin, Stranski-Laboratorium,Strasse des 17.Juni 112, D-10623 Berlin — [2]Universität Ulm, Abteilung Angewandte Physik, D-89069 Ulm — [3]MPI-KGF, Potsdam-Golm,14424 Potsdam

The bulk swelling behavior of copolymer microgel particles is compared to the swelling behavior of adsorbed particles. The transition temperature of these microgel particles is found to be ≈ 32°C. Microgel particles adsorbed onto a solid substrate still display a similar reversible volume phase transition as their dissolved counterparts. However, their swelling ratio α is reduced by approximately one order of magnitude compared to the bulk value. Nevertheless, the effect is sufficiently large to make the particles suitable for applications as sensors and actuators.
In addition first results for microgel layers embedded in polyelectrolyte multilayers are presented. These new materials are promising with respect to senosric applications.

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Closing remarks — •Erwin Frey —