BP 38: Focus: Charge Effects in Soft and Biological Matter III (joint CPP, BP, ST)

Time: Friday 10:15–12:00   Location: H45

Invited Talk   BP 38.1   Fri 10:15   H45
Charge inversion in macromolecular systems — •Christian Holm — Institut für Computerphysik, Universität Stuttgart, Pfaffenwaldring 27, 70569 Stuttgart, Germany

We discuss our current understanding of the phenomenon of charge inversion in macromolecular systems, based on simulational results in recent years. In this approach the solvent has been incorporated only as an implicit dielectric background, whereas all charges and salt ions are treated explicitly. We will also discuss our recent work on modelling electrophoresis of charged polymers within a coarse grained approach, where the solvent degrees of freedom have been modelled using a Lattice-Boltzmann algorithm.

[1] R. Messina, C. Holm, K. Kremer, *Ground state of two unlike charged colloids: an analogy with ionic bonding*, Euro. Phys. Lett., **51**, 461 (2000).
[2] O. Lenz, C. Holm, *Simulation of charge reversal in salty environments: Giant overcharging?*, Euro. Phys. J. E **26**, 191-195 (2008).
[3] K. Grass, C. Holm *Mesoscale modelling of polyelectrolyte electrophoresis*, Faraday Discuss. **144**, 57-70 (2010).

                                                BP 38.2   Fri 10:45   H45
Counterion condensation and effective charge of linear and globular macromolecules — Ute Böhme and •Ulrich Scheler — Leibniz Institute of Polymer Research Dresden, Hohe Strasse 6, 01069 Dresden

The charge density on charged macromolecules is usually so high, that the thermal energy of the respective counterions is insufficient to escape the electric field generated from the charges on the macromolecule. Therefore a fraction of counterions condenses on the macromolecule, lowering the effective charge of the macromolecule The combination of diffusion and electrophoresis NMR provides an umambiquous possibility for the experimental determination of the effective charge, which is in good agreement with molecular simulations [1, 2]. This approach has been applied to flexible and stiff polyelectrolytes as well as proteins and other globular molecules [5]. Because PAMAM dendrimers exhibit only two types of chargeable groups, counterion condensation can easily be quantified, where the degree of protonation of the amino groups is inferred from proton NMR spectra. The fraction of condensed counterions increases with increasing molecular weight to level at about 70%. The effect of the variation of the solvent properties is studied with variation of the ionic strength and the dielectric constant of the solution.

[1] K. Grass, U. Böhme, U. Scheler, H. Cottet, C. Holm, Physical Review Letters 100, (2008) 096104 [2] U. Scheler, Current opinion in Colloid & Interface Science, 14 (2009), 212 [3] U. Böhme, U.Scheler, Coll. & Surf.A„ 222, (2003), 35

                                                BP 38.3   Fri 11:00   H45
Detection of multilayer formation of charged macromolecules by field-effect devices: from experiment to theory — •Andrey Cherstvy[1], Mariam Abouzar[2], and Arshak Poghossian[2] — [1]IFF-2, FZ Jülich, Germany — [2]IBN-2, FZ Jülich, Germany

Field-effect based semiconductor devices for label-free detection of adsorption of charged macromolecules are widely used for biosensor applications. The quantitative understanding of signals measured is however still missing in many cases. We use a capacitive electrolyte-insulator field-effect device for electrical detection of layer-by-layer adsorption of oppositely charged polyelectrolyte PE PSS/PAH chains onto the sensor surface [1]. We measure the thickness of PE layers formed as well as the changes in morphology and wetability of the modified sensor surface. We also study the effects of ionic strength on the sensor signal detected upon PE multilayer formation. We observe progressively decaying oscillations of the sensor potential upon repetitive PE adsorption. To analyze these zig-zag variations, we develop a theoretical model that accounts for the Debye screening by mobile ions within the PE layer. The model predicts that potential oscillations monitored by the sensor originate from the sign and the charge density of last PE layer. These variations depend strongly on bulk electrolyte concentration and physical parameters of PE multilayers. At lower salt, the oscillations predicted are much larger and more

persistent with number of PE layers deposited. The magnitude and decay length of oscillations are in good agreement with experimental observations.

[1] A. G. Cherstvy et al., submitted to J. Phys. Chem. B.

                                                BP 38.4   Fri 11:15   H45
Electrostatic interactions control the permeability of biological hydrogel filters — •Oliver Lieleg[1,3], Lucy Colwell[1,2], Regina Baumgärtel[1,3], Ioana Vladescu[1], Michael Brenner[2], Andreas Bausch[3], and Katharina Ribbeck[1] — [1]FAS Center for Systems Biology, Harvard University, Cambridge, USA — [2]School of Engineering and Applied Sciences, Harvard University, Cambridge, USA — [3]Lehrstuhl für Zellbiophysik E27, Technische Universität München, Germany

The controlled exchange of molecules between biological entities (organelles, cells, or organisms) and their environment is critical for life. Biological hydrogels appear well suited to achieve such selective exchange: A hydrogel within the nuclear pore controls the passage of material between the nucleus and the cytoplasm. Mucus hydrogels lining the uterus, the stomach or the lung allow us to expel ingested particles and defend the cells beneath from a variety of pathogens. Extracellular matrix hydrogels in the connective tissue regulate the distribution of growth factors, proteins or drugs. Although hydrogel based filters are integral parts of biology, clear concepts of how their barrier function is controlled on a microscopic level are missing. Here, we discuss three biological hydrogels which differ in terms of their composition and biological function, but nevertheless seem to share a common physical design principle that regulates their microscopic barrier function: We demonstrate that particle translocation in these hydrogels is based on electrostatic interactions between diffusing particles and the hydrogel polymers rather than size exclusion effects.

                                                BP 38.5   Fri 11:30   H45
Diffusion and charge transport in ionic liquids: the role of ion-ion interactions — •Joshua Sangoro, Ciprian Iacob, Jörg Kärger, and Friedrich Kremer — Institute of Experimental Physics I, University of Leipzig, Linnéstr. 5, 04103, Leipzig

Self-diffusion in a variety of glass-forming ionic liquids (ILs) are investigated in a wide frequency and temperature range by means of Broadband Dielectric Spectroscopy(BDS) and Pulsed Field Gradient Nuclear Magnetic Resonance (PFG NMR). It is experimentally shown that in the time-scale characterising the cross-over from sub-diffusive to diffusive ion dynamics, the hopping lengths are of the order of molecular diameters determined from quantum-chemical calculations. This provides a direct means - via Einstein-Smoluchowski relation - to determine diffusion coefficients by BDS over more than 8 decades unambiguously and in quantitative agreement with independent PFG NMR measurements. Unprecedented possibilities in the study of charge transport and dynamic glass transition are thus opened.

                                                BP 38.6   Fri 11:45   H45
Collapse of highly charged polyelectrolytes triggered by attractive dipole-dipole interactions — •Andrey Cherstvy — IFF-2, FZ Juelich

We study the collapse of flexible highly charged polyelectrolyte PE chains induced by attractive dipole-dipole interactions [1]. The latter emerge due to formation of dipoles between the chain monomers and counterions condensed on PE from solution. Using the statistics of slightly perturbed Gaussian polymers, we obtain the scaling relations for PE dimensions as a function of PE charge density in the limit of compacting chains. The results are in good agreement with the outcomes of MD simulations of collapse of flexible PEs with explicit counterions. Then, we analyze the results of MD simulations for complex formation by two highly charged PEs carrying opposite charges. We use the scaling arguments based on the picture of complexation of electrostatic blobs to rationalize the size of complexes of the two chains in the collapsed state. We also analyze PE linear charge densities required for the onset onto collapse. [1] A. Cherstvy, submitted to JPCB.