

Proposal within the SPP 2171:
Dynamical Wetting Behavior of Flexible Charged Substrates

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Project Description

1 State of the art and preliminary work of the applicants

Brief Introduction

Electrowetting (EW) is an indispensable and versatile means for droplet manipulation and control of surface wetting. It is perhaps the most flexible method to actively control the wetting behavior of conductive liquids on partially wetting surfaces. EW has applications in a range of applications from silicon devices to optic lenses, and researchers from various disciplines try to study the unique properties of EW. A thorough understanding of the fundamental properties of materials and liquids that lead to dynamic wetting is, however, still missing. Moreover, how an applied electric field, the pH of the solvent, or characteristics of the surface may influence the shape, orientation or contact angle saturation [1, 2] of the droplet has not been entirely understood, thereby igniting much research efforts in this area [1, 3–5].

The group of one of our collaborators [6] has recently shown that the Gibbs transfer energy represents a novel control parameter of the wetting properties of drops as it can be tuned by modifying the concentration of water in aqueous electrolyte solutions [7, 8]. Use of advanced mathematical models and computer simulations is indispensable for an understanding of phenomena that cannot otherwise be predicted with experiments. Collaboration of experimentalists, theorists, and computational scientists thus seems to be the path to further our understanding in this area.

In this project we propose to develop a computational model capable of simulating an aqueous droplet sitting on adaptive, flexible charged surfaces formed by charged macromolecules. These surfaces are responsive to electric field and pH stimuli, and the supporting surface could also have topographic features like charge patchiness, variable hydrophobicity/hydrophilicity or corrugations. We want to study the effects that these stimuli have on the surface properties, and hence of the wetting behavior, and how they dynamically interact. We will validate our model with experimental data from collaborators within the SPP.

Below we briefly outline the current state of research in this area, our own preliminary work, and the achievements we hope to gain in this field.

Experiments

A number of research groups have been conducting experiments to study EW of different kinds of surfaces as it has a wide range of applications including lab-on-chip devices, lenses and display screens. Experiments have reached a stage of maturity in which a number of parameters and physical processes that have influence on EW can be investigated. Researchers consider *contact angle hysteresis* [9, 10] to be one of the most interesting characteristics of study in these applications, and try to explore how properties of the surface [11] and applied voltage may influence the contact angles [12].

Early experimentalists used thick insulating layers and demonstrated a contact angle saturation mechanism that involved the ejection of satellite droplets from the contact line [13, 14]. Vallet et al. [13] proposed that the diverging charge density at the three phase contact line leads to the excitation of transverse modulations of the contact line, which eventually become *unstable* beyond a certain applied voltage. A quantitative analysis is still however lacking as an analysis of electric field distribution in a three-dimensional geometry involves complexity. Further challenges include contact angle hysteresis and contact line pinning due to surface heterogeneity as reviewed by De Gennes [15].

A range of fluids, fluid and solvent mixtures as well as immiscible fluids are studied for an insight into EW. Recent work of Gao et al. [16] is exemplary in the direction of immiscible fluids where the authors demonstrated that increasing voltage, both AC or DC, increased the contact angle hysteresis in ambient oil compared to the case of ambient air. They found this effect to be caused by the fact that the oil film under the droplet decreases in thickness under the influence of the Maxwell stress and thus loses its ability to smoothen the intrinsic substrate roughness and to lubricate the contact line. The same group [17] also demonstrated the influence of structured electrodes on the distribution and shape of droplets.

It has been recently shown by Sénéchal et al. [18] how the effective ionisation degree of the grafted weak poly-acid can be controlled with the externally applied field and environmental conditions namely pH and ionic strength. Chevalliot et al. [2] have demonstrated that when using DC voltage, EW contact angle saturation is independent of electric field, contact line profile, inter-facial tension, choice of non-polar insulating fluid, and type of polar conductive fluid or ionic content.

While the influence of the electric field and solvent pH on surface EW properties is important to study experimentally, the characteristics of the surface itself have also been of interest. For example recent works of Papathanasiou [19] and Yurkiv et al. [20] focus on superhydrophobic surfaces while Dai and Li [21] developed a EW module with a hydrophilic grid by forming a patterned layer of electrodes on a substrate.

Computational modeling

A number of research groups use computational modeling to answer research questions about EW. Papathanasiou and co-workers [19, 22, 23] have conducted a number of numerical and theoretical studies ranging from investigations of electric fields close to contact line to voltage dependent changes in droplet shape. Models of Peykov et al. [24] and Quinn et al. [25] are also worth noting as they are easy to apply and obtain saturation angles and voltages in good agreement with experiments. Computational Fluid Dynamics (CFD) studies have also investigated the basic processes of drop motion, its splitting and control [26, 27] only to find *qualitatively* good agreements with experiments while *quantitative* agreements are mostly based on empirical correction factors accounting for the unknown contact line friction.

A range of numerical methods are used in such studies including Lattice Boltzmann Method (LBM), phase field methods and molecular dynamics or a combination of these. Here we discuss the current state of research in this area based on the employed numerical methods.

Use of Molecular Dynamics in modeling various polyelectrolyte architectures or gels

The first simulations using simple bead-spring models for polyelectrolytes (PEs) have been performed by Stevens and Kremer [28, 29]. Extensive reviews about the simulations and the theory of polyelectrolytes can be found in Refs. Dobrynin and Rubinstein [30], Messina [31], and Košovan et al. [32]. Uncharged polymer brushes have been extensively modeled by various research groups using coarse-grained molecular dynamics models. Binder et al. [33] provides an eloquent review of MD simulations to model polymer brushes. Previous works of Grest [34, 35] and Kremer and Grest [36] are the most exemplary in this direction as they explore the tribological properties of surfaces that are coated with brushes. More recently Galuschko et al. [37] computed frictional forces between non-entangled polymer brushes using a coarse-grained model and found excellent agreements with theory, while Singh et al. [38, 39] used similar methodology to find good agreement with experiments. The advantage of using a coarse-grained description of polymers for the research questions addressed in the aforementioned literature lies in the spatial and temporal scales that can be attained at moderate computational costs, without the need for high performance computing resources. The polymer brushes in these works are modeled as a set of semi-flexible, multibead-spring chains with excluded volume, where each chain is permanently fixed at one of its ends at random position on one of the two opposing surfaces. Recent works have also focused on the effect of solvent conditions on PE brushes [40] while others [41] model effect of electric fields on these brushes. A recent work of De Beer et al. [42] used MD and found that two immiscible brush systems in mechanical contact slide along a fluid-fluid interface while having load-bearing ability.

Use of Lattice Boltzmann Method in modeling solutes, electrolytes and multiphase flows

The lattice Boltzmann (LB) method is a popular mesoscopic approach for simulating hydrodynamic interactions in complex fluids [43]. The solvent is represented by a discrete phase space distribution of particle populations residing on lattice nodes. These while undergoing local collisions, changes in populations are propagated to nearby nodes. By means of multi-scale Chapman-Enskog expansion of the Boltzmann transport equation, it can be shown that on sufficiently large length and time scales, these microdynamics lead to hydrodynamic flow that satisfies the Navier-Stokes equations. The LB method can thus be used as a computationally efficient tool for hydrodynamics. Furthermore, the method can be generalised for multicomponent flows and readily allows for an efficient coupling with other methods. For example, using Molecular dynamics to couple dispersed particles to a fluid.

The Shan-Chen (SC) method [44] allows for the simulation of immiscible binary fluids by introducing a phenomenological repulsive interaction potential between two LB fluids. The advantage of the SC approach over others lies in its simplicity as it allows one to carry out local calculations efficiently and to easily incorporate other relevant physical effects into one SC framework. A critical review of various LB algorithms for the simulation of multicomponent fluids have found the SC approach one of the most suitable [43].

Furthermore, this algorithm allows for a straight-forward way of tuning the wettability of the fluids with respect to the solid boundaries by introducing a virtual fluid inside the solid nodes. This virtual fluid consists of a tuneable ratio of the two real fluid components, leading to preferential wetting of one of the fluid components upon boundary contact. This technique has been successfully applied, for example by Davies et al. [45] who studied the adsorption of particles at fluid-fluid interfaces.

An arbitrary number of miscible (possibly ionic) chemical species can be coupled to the LB fluid by means of the electrokinetics (EK) scheme first introduced by Capuani et al. [46], and in the forthcoming sections always abbreviated as LB-EK. This method solves an advection-diffusion equation for each miscible species using a finite volume scheme and allows for definition of arbitrary chemical potentials. It has been used to simulate electroosmotic flow and sedimentation of charged particles [46], electrophoresis of individual colloids [47] and suspensions thereof [48]. Recently, this method has been extended to include moving boundaries of charged colloids, which extends its application to

electrophoresis applications of colloids and charged model proteins [48]. This combination of the LB-EK method allows for the efficient and large-scale simulation of multi-phase, multi-component flow, as will be necessary in this investigation. Furthermore, recently Rivas et al. [49] have presented a two-phase LB-EK model, which also takes the ion partitioning between the two phases and moving boundaries of colloidal objects into account.

Coupling between the LB fluid and particle-based molecular dynamics (MD) can be achieved using a number of different methods, depending on the required resolution and the nature of the immersed particles. If the structure of these particles and details of the flow in their vicinity are of importance, the moving boundary method [50] or the immersed boundary method [51] can be used. Both of these methods exchange momentum between the (discretised) particle surface and the fluid. The moving boundary method achieves this via a bounce-back scheme (inverting the direction of propagation of LB populations entering the boundary), while a number of different coupling schemes based on forces and bounce-back exist in the case of the immersed boundary method. The immersed boundary method is most often used for objects that are flexible [52], with biological cells as notable example; the moving boundary method on the other hand is most often used for rigid objects [48], such as colloids or other macroscopic bodies. If one is interested in (longer range) hydrodynamic interactions between particles or between particles and surfaces, and the specific shape of individual particles and the surrounding flow is less of an issue, one can employ the computationally more efficient point coupling, in which a point force derived from Stokes' law acts between fluid and particle [50]. This type of coupling leads to a coarser description of the system in which MD particles obtain an effective hydrodynamic radius by means of the grid resolution and their friction coefficient. Despite its simplicity, this scheme can correctly reproduce effects as diverse as the dynamics of polyelectrolytes, the concentration dependent conductivity of electrolytes, particle-wall interactions, and the dynamics of self-propelled particles [48, 53].

Own preliminary work of the applicants

PI Holm has more than twenty years of experience with the study of charged and magnetic soft matter systems. These are either treated theoretically with mean-field approaches like the Poisson-Boltzmann theory or the standard electrokinetic continuum equations. Due to the difficulty of treating long range interactions analytically, we mostly use molecular dynamics and Monte-Carlo methods to investigate charged and magnetic soft matter, as well as their hydrodynamic interactions. For the study of dynamical quantities we have been using suitably modified lattice-Boltzmann algorithms with the Dünweg-Ahlrichs[54] point coupling scheme in the past. Since 2001 we have been the main developer of the ESPResSo [55, 56] suite which consists of various modules ranging from MD, LB to DPD to study soft matter systems. We have developed an efficient and parallel GPU-based LB solver for the continuum electrokinetic equations based on the approach by Capuani et al. [46], but so far only for a single phase LB model. We have also shown recently how to significantly improve the stability of the EK-LB scheme by reducing spurious flux contributions [57]. PI Holm has ample experience with electrolyte flow and charged macromolecules in general, and with polyelectrolyte multilayers[58, 59], pH-sensitive polyelectrolytes[60, 61] and charged hydrogels [32, 62]. For example, we have studied in the past the stretching of charged macromolecules in the presence of electric fields and flows[63, 64]. We also recently started to implement algorithms that allow us to treat charged interfaces and charged macromolecules with pH-dependent surface charges, meaning that they undergo charge regulation using a Monte-Carlo method called the reaction ensemble [65, 66]. We have successfully collaborated with other SPP groups in the past [59] on related projects. For large-scale systems and complicated geometries requiring very high resolution, we additionally employ the high performance code waLBerla that scales efficiently up to millions of cores ([67]), and into which we have also implemented the single flow EK-LB with appropriate moving boundary fluid coupling to particles [48].

PI Jain has more than 7 years of experience with LB based hydrodynamics. His research has focused on various aspects of multi-scale modeling, fluid dynamics, high performance computing, transport

in porous media, epithelial and paracellular flow models, microvasculature, and biomedical fluid dynamics. He is one of the developers and main contributors to the *Musubi* [68, 69] LBM solver, which is part of the end-to-end massively parallel simulation framework APES [70, 71]. *Musubi* is known to exhibit one of the best efficiency and parallel scalability on massively parallel supercomputers as evidenced by literature [71, 72]. *Musubi* and *waLBerla* in fact are the only LB solvers showcased by the Forschungszentrum Jülich in their High-Q club as one of the very few codes that scaled about a million threads of the whole machine¹. *Musubi* implements Maxwell-Stefan diffusion of electrolytes, passive scalar transport [73] and has been used for answering complex questions in Physiological applications [72, 74]. PI Jain has used and enabled scaling of this solver on major Tier-0 Supercomputers in GERMANY, JAPAN and SWITZERLAND and various suites within the APES framework are used by many research groups across the globe.

1.1 Project-related publications

1.1.1 Articles published by outlets with scientific quality assurance, book publications, and works accepted for publication but not yet published.

- [P1] H. J. Limbach, A. Arnold, B. A. Mann, C. Holm, “[ESPResSo – An Extensible Simulation Package for Research on Soft Matter Systems](#)”, Comput. Phys. Commun. **2006**, 174, 704–727
- [P2] G. Rempfer, G. B. Davies, C. Holm, J. de Graaf, “[Reducing spurious flow in simulations of electrokinetic phenomena](#)”, J. Chem. Phys. **2016**, 145, 044901
- [P3] D. Sean, J. Landsgesell, C. Holm, “[Computer Simulations of Static and Dynamical Properties of Weak Polyelectrolyte Nanogels in Salty Solutions](#)”, Gels **2018**, 4, 2
- [P4] J. Landsgesell, C. Holm, J. Smiatek, “[Wang-Landau Reaction Ensemble Method: Simulation of Weak Polyelectrolytes and General Acid-Base Reactions](#)”, J. Chem. Theor. Comput. **2017**, 13, 852–862
- [P5] S. Micciulla, P. A. Sanchez, J. Smiatek, B. Qiao, M. Sega, A. Laschewsky, C. Holm, R. von Klitzing, “[Layer-by-layer formation of oligoelectrolyte multilayers: a combined experimental and computational study](#)”, Soft Mater. **2014**, 12, S14
- [P6] M. Kuron, G. Rempfer, F. Schornbaum, M. Bauer, C. Godenschwager, C. Holm, J. de Graaf, “[Moving charged particles in lattice Boltzmann-based electrokinetics](#)”, J. Chem. Phys. **2016**, 145, 214102
- [P7] K. Jain, S. Roller, K.-A. Mardal, “[Transitional flow in intracranial aneurysms—a space and time refinement study below the Kolmogorov scales using Lattice Boltzmann Method](#)”, Comput. & Fluids **2016**, 127, 36–46
- [P8] J. Qi, K. Jain, H. Klimach, S. Roller in Advances in Parallel Computing: On the Road to Exascale, IOS Press, **2016**, pp. 807–816
- [P9] K. Jain, G. Ringstad, P.-K. Eide, K.-A. Mardal, “[Direct numerical simulation of transitional hydrodynamics of the cerebrospinal fluid in Chiari I malformation: The role of crano-vertebral junction](#)”, Int. J. for Numer. Methods Biomed. Eng. **2017**, 33, e02853

1.1.2 Other publications

- [P10] F. Weik, R. Weeber, K. Szuttor, K. Breitsprecher, J. de Graaf, M. Kuron, J. Landsgesell, H. Menke, D. Sean, C. Holm, “[ESPResSo 4.0 – An Extensible Software Package for Simulating Soft Matter Systems](#)”, arXiv:181107729 **2018**

¹http://www.fz-juelich.de/ias/jsc/EN/Expertise/High-Q-Club/_node.html

1.1.3 Patents

EP2447867 - Method for determining the performances of a superabsorbent polymer material, together with P. Verstraete, T. Lindner, A. Meyer, M. Schmidt, K. Grass. (Procter & Gamble)

2 Objectives and work programme

2.1 Anticipated total duration of the project

The initial phase of the project is expected to run for three years of the Schwerpunktprogramm 2171, starting from 01 Oct 2019 through 30 Sep 2022.

2.2 Objectives

Our objectives are to study the dynamic wetting properties of adaptive, flexible, charged surfaces that are made of partially charged macromolecules in contact with aqueous droplets of different electrolyte concentrations. The macromolecular surfaces can be thin films composed of linear polyelectrolytes, polyelectrolyte multilayers, polyelectrolyte brushes, microgels, macrogels, nano-grass, or other complicated macromolecular structures like amphiphilic co-networks. The solid surface supporting the thin film of macromolecules itself can also be nano-structured, in terms of preferentially hydrophobic or hydrophilic domains, or be a surface corrugation on the nanometer scale. The switching of surface properties can be induced by various methods. We plan to study the effects of electrical fields and a varying pH-value of the aqueous solution as our control parameter on the wetting dynamics. Temperature will not be a control parameter, but we still can study thermoresponsive hydrogels via an effective parametrisation of the polymer interactions.

Our method combines continuum approaches like the solution of the system of electrokinetic equations with a responsive fluid-structure coupling of charged macromolecules. The continuum electrokinetics will be modeled via the in Sec. 1 described lattice-Boltzmann based electrokinetic (LB-EK) solver. This solver exist as a single phase fast GPU implementation in our software package ESPResSo. We know how to couple fixed dielectric boundaries and moving boundaries to the LB-EK [48, 57], as is useful for charged colloidal sized objects. However, the question of how to couple charged polymers to the LB-EK such that they retain their correct electrostatic interactions, hence their scaling properties, is an unsolved problem. To study the water in contact with air and a solid, we will use the extension of the LB-EK to a Shan-Chen multiphase flow model. This has already been suggested by Rotenberg et al. [76] and investigated by the group of Harting [6, 49]. The inclusion of pH-dependant effects has to be done in a two-fold way. In the LB-EK, a change in pH is modeled via a suitable boundary conditions, like is routinely done in the Poisson-Boltzmann equation. On the other hand, the charge fluctuations and dissociation reactions on the polymer beads can be taken into account via the reaction ensemble Monte Carlo method [65]. As external stimuli we will apply DC or AC electric fields that change the ion distribution, and hence have an influence on the charge regulation. Moreover they will also change the behavior of the functionalised surface, since they can swell or deswell, or to be more or less wettable. We expect that the change in surface properties will also give rise to interesting effects in the wetting dynamics.

A 2D schematic picture of the proposed system is shown in Figure 1, displaying the computational components of the model. The charged polymers lie at the interface of the droplet and the surface (see figure 1) and will be *explicitly* modeled through a particle coupling approach where we will use molecular dynamics (MD) for the dynamics of the beads. We stress here that the coupling of **charged** macromolecules to a multiphase LB-EK with a yet an unexplored option, which would require

development of a suitable coupling scheme and validation thereof. An explicit modeling of the polymers at surfaces would pave the way for future studies using patterned mobile and responsive surfaces such as nano-grass, cilia or flaps to name a few. Going even further, the creation of a LB-EK scheme with a particle coupling to explicitly charged polymers has the potential for study of a number of other areas like electrokinetic drug release with nanogels, or the study of protein and DNA dynamics in electrolyte solutions of varying pH conditions.

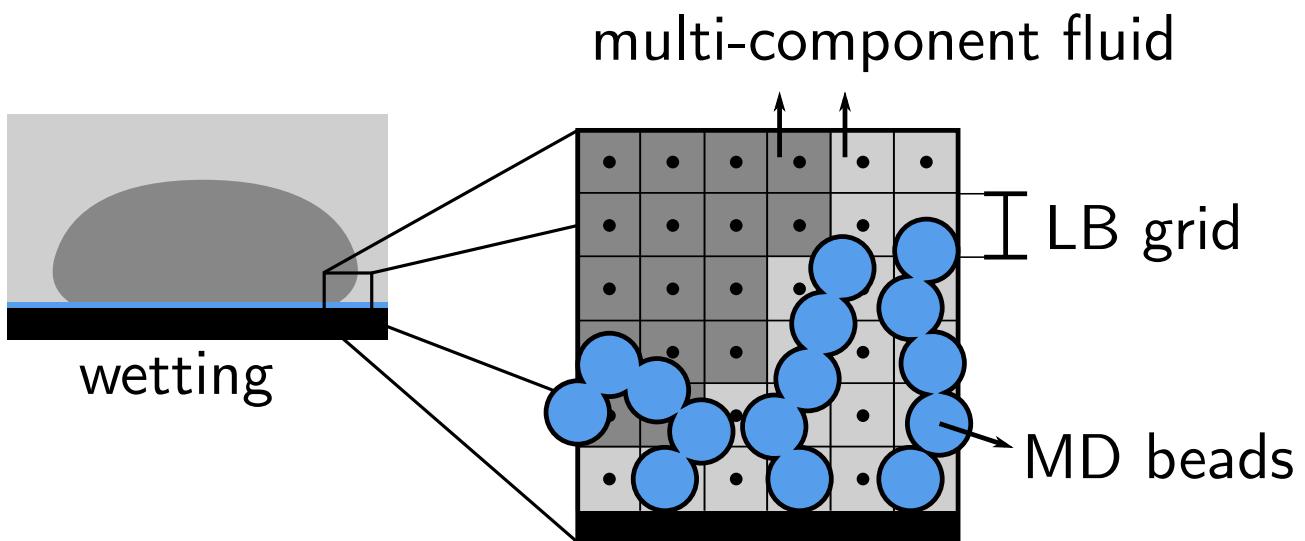


Figure 1 Schematic of the simulation system. The dynamics of a multi-component LB fluid with an implicit electrokinetics solver is computed on a simple cartesian grid. This charged multiphase fluid is in turn coupled to charged off-lattice MD beads making up several polyelectrolytes. The polyelectrolytes can also be crosslinked to represent a microgel, or polyelectrolytes with alternating charges can form polyelectrolyte multilayers. The so-formed flexible surface on which an aqueous droplet can be put, can be hydrophobic or hydrophilic, and can even be patchy with regions of hydrophobic and hydrophilic material. Applied changes in the solution pH or electric fields will change the substrate dynamics as well the wetting properties.

2.3 Work programme including proposed research methods

Our research proposal is partitioned into 4 work packages (WPs) that target the specific problems that we want to address in our SPP proposal: In WP1 we start with the implementation of a LB-EK Shan-Chen multiphase model capable to respond to solution pH changes, whose implementation can be validated with experiments performed in the groups of **Pachauri** and **Huber**. In WP2 we focus on the development of a coupling scheme of explicitly charged polymers to the LB-EK scheme that can also respond to pH and electric field changes, opening up the ability to study the mutual responses of flexible charged substrates to an aqueous wetting fluid under electric field and solution pH changes. In WP3 we will apply our model to systems to a polyelectrolyte multilayer system as studied experimentally by **von Klitzing** and on the atomistic scale by **Sega**. After having established the methods for some experimentally investigated systems we can in WP4 look at other flexible substrates made up of charged macromolecules like polyelectrolyte brushes, hydrogels, microgels to make contact to several other experimental groups like **Müller** or **Monkman**. We intend to look at more complicated scenarios, where we will also incorporate hydrophobic elements into the polymer, opening up the way for micro-phase separated polymeric structures that can form hydrophilic and hydrophobic domains under suitable external stimuli that should give rise to a host of interesting wetting phenomena.

WP1: Implementation of a two-phase Shan-Chen multiphase electrokinetics solver working with fixed charged boundaries

The first workpackage WP1 consists of the implementation of a two-phase Shan-Chen model with the electrokinetics solver. A viable route to do this has been presented recently by Harting [49]. We

will start out by implementing a Shan-Chen model into our GPU-based LB that is compatible with our EK-LB. The substrate will in this first part be a simple flat wall with different wetting properties, like in the experiments of the **Pachauri** group, or a regular porous sample with nanochannels standing perpendicular to a substrate, like in the experiments of the group of **Huber**. This part will be followed by the inclusion of pH-sensitive walls into the EK-LB on the basis of the equivalent Poisson-Boltzmann treatment. This means that the surface charge densities are not fixed, but the degree of dissociation depends on the local field strength, hence depends on the ion distributions, solution pH, as well as the flow velocity. Finally, we will proceed to implement the most suitable model for the treatment of high-density-difference two-phase flow (water and vapour/air). This model can now be parametrised and validated with the experiments performed in the groups of **Huber** and **Pachauri**. We can perform measurements of the wetting angles, the water imbibition, the conductivity, the capacitance, and the change of wetting angles if we change the surfaces from hydrophilic to hydrophobic. These comparisons should give us a mapping from model parameters to real-world systems, and reveal possible shortcomings of the implementation or the model. We can also apply an electric field to probe electrowetting properties [6], or change the pH-value to see equivalent changes in wetting behavior. The incorporation of flexible surfaces will be left to the next working package.

WP2: Development of a coupling method for charged flexible polyelectrolytes to the LB-EK

In this working package we will develop a particle coupling scheme by which we can link flexible bead-spring charged polymer models to the LB-EK (see figure 1). We know how to couple stationary charged boundaries to the LB-EK, and we have developed a moving boundary coupling scheme to LB-EK [48, 49]. However, in order to couple flexible polymers fast to an LB-EK we would like to apply a fast point-coupling scheme that has been successfully done for uncharged polymers to the LB by Dünweg and Ahlrichs[77]. A simple moving boundary coupling would be simply to costly since we would need a very fine grid resolution to resolve the particle sufficiently. We have started preliminary work on this coupling scheme in the Bachelor thesis of S. Schöll, (ICP, unpublished). In Figure 2 we demonstrate that for fixed boundary conditions, where the polymer is modelled as a series of charged beads, see 2a, or a charged fixed rod with a certain radius that the ions can not penetrate, we obtain very good agreement with the Poisson-Boltzmann theory, as it should be, see figure 2b. We propose to develop a model that would represent the polymer as flexible combination of charged particles with a density of ions that will be determined via the LB-EK, as depicted in the right panel, see figure 2c.

We have found that a simple point-coupling approach according to Ref. [77] does not yield the correct inter-chain interactions of charged beads, see also [78]. Therefore we need to explore this coupling scheme more extensively and add suitable modifications. We intend to test an interpolation scheme to exclude ions around the charged MD beads, and we will also try to thermalise the LB-EK consistently, since for consistency both parts of the algorithm, the MD part and the lattice part, should be thermalised on equal footings. The to be developed algorithm will require extensive validation and verification against explicit ion simulations, which will serve as reference calculations. This can be done for static properties like the scaling of the radius of gyration as a function of the degree of polymerisation and salt concentration, but also we intend to test the dynamic properties like the electrophoretic mobility as function of chain length and salt concentration. Here we can compare to our old data on electrophoretic mobilities[79, 80].

WP3: Application of the particle-coupled LB-EK to flexible charged substrates: Case study PEM

In the beginning we would like to apply the particle coupling to polyelectrolyte-multilayers (PEMs). These consist of alternating layers of positively and negatively charged polyelectrolytes and have been studied by us via atomistic and coarse-grained simulations [59, 81, 82] before. From the atomistic reference conformations we extract a coarse-grained representation which we want to study with the LB-EK. Water droplets of varying electrolyte concentration will be used to measure contact angles, and

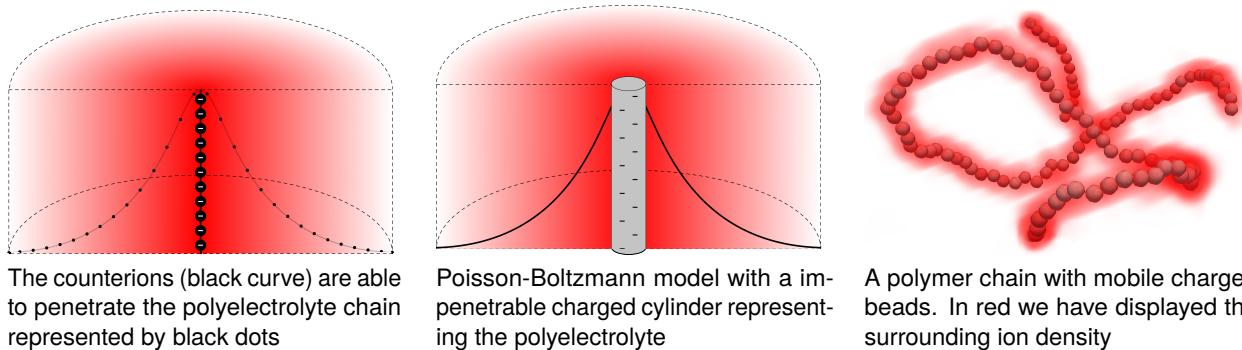


Figure 2 Our previous work on coupling a polyelectrolyte chain to the LB-EK. The middle figure shows the results of a Poisson-Boltzmann model where the charged cylinder that represents a polyelectrolyte is impenetrable. The right images represents a polymer chain with charged beads and a surrounding ion density.

deformation of the droplet under an applied force (here gravity in form of a tilted substrate). Exactly this scenario will also be investigated experimentally in the group of **R. von Klitzing** and via atomistic simulations in the group of **Sega** which whom we can compare our results. We plan to have either the upper surface to be positively charged or negatively charged. Since we plan to use only a low numer of multilayers (up to four), the properties of the underlying surface will most likely still have an effect on the structure of the multilayer, and hence also on the wetting properties. Therefore we do not expect symmetric wetting profiles. In addition we plan to look at changes of he wetting behavior as a function of pH value. The bottom surface, on which the PEM resides, is normally a glas substrate that is pH-dependant. Any change in the solvent pH would also affect the charge of the glass, that then again effects the conformation of the PEMs, and hence reacts back on the wetting properties. We will also study the effects on an applied electric field normal to the surface on the wetting properties.

WP4: Incorporation of hydrophobic elements in the flexible charged substrates

The last working package is basically open ended, since, if all our previous WPs are worked out, we can start of building more complicated flexible substrates. The surface may be covered with other polymer topologies like brushes, dendrimers, microgels, or crosslinked hydrogels, etc, which should all carry charges. Moreover, the surfaces can be hydrophobic or hydrophilic, which will have an effect of the surface properties, that might also effect the topmost layer. Moreover, we can add hydrophobic polymer blocks to the charged hydrophilic blocks, such that the microstructure of the macromolecular surface layer can develop an interesting microstructure, namely hydrophilic and hydrophobic domains. From simple linear polymers we know that theses systems can micro-phase separate into pearl-necklace conformations [83–85]. These substrate restructuring can have interesting new effects on the wetting behavior, and again could show an interesting surface-wetting coupling. Changes in the properties and structure of the hydrophilic- hydrophobic domains can be induced via electric fields or changes in the pH, both means that will be accessible via our developed algorithms. Here we plan extensive parameter studies to map out interesting regimes. Here we could benefit from additional experiments performed in the groups of **von Klitzing** and **Müller**. Obvious questions that could be answered are: How does the applied electric field change the wetting properties of the surface, and do AC and DC electric fields yield fundamentally different changes? How does the droplet contact angle changes as a function of electrolyte concentration and electric field? Do AC or DC fields reduce or enlarge the contact angle? How does the solvent pH influence the wetting properties and how does this compare to the aforementioned changes being brought by the electric field? Moreover we can modify the surface on which the flexible substrate is suspended. What does a change in corrugation yield, what do hydrophilic or hydrophobic patches do to the overall surface layer, or how will all this affect the overall wetting properties. Here we can make contact to the experiments on uncharged layers from the **Rühe** group. All these are important questions that we would like to address if our research time permits this.

Computational Implementation:

We plan to implement all the code required for the simulations proposed in the context of this proposal in the Stuttgart-maintained software package ESPResSo [55, 56], which was described before. In addition we have an ongoing collaboration with the group in Erlangen that developed and maintains the waLBerla [67] software package. ESPResSo offers principal advantages in the context of this proposal because of the availability of an MD solver and a single phase LB-EK scheme implemented within the same package [86]. waLBerla, on the other hand, provides an excellent scaling on supercomputers and might be useful for the study of very large systems. We have plans for the future to interface waLBerla to ESPResSo in order to be able to study very large systems for the cases where very fine grid resolutions are needed.

Outlook for the next funding period

Having implemented the necessary tools for studying the dynamic aspects of wetting on flexible charged substrates will open up the door to study a host of interesting substrates and their response to pH changes and applied electric fields. In principle we can also incorporate magnetic field responses to magnetically interacting inclusions into the substrate, to study their interfacial dynamics, and other effects such as capillary attractions between immersed particles, or even more complicated substrate compositions. The proposed investigations in WP4 will probably not have been finished within the proposed three year period, and we foresee that many more challenging problems will surface over time, which can then be investigated in a subsequent funding period. We can also make contacts to other interesting experiments that will certainly be performed within the SPP, and that we are currently not aware of.

Planned schedule

The following time table gives rough estimates of the research time that the different parts of the work plan require and how they are spaced within the funding period. We indicate the quarters of a year using the numbers 1–4 and the color bar slides according to our plan schedule for each WP:

		Year 1				Year 2				Year 3			
		1	2	3	4	1	2	3	4	1	2	3	4
WP1	Two phase Shan-Chen EK solver with fixed charged boundaries												
WP2	Coupling method for charged flexible polyelectrolytes to the LB-EK												
WP3	Application of the particle coupled LB-EK to flexible charged substrates												
WP4	Hydrophobic elements in the flexible charged substrates												

2.4 Data handling

The processed and analysed data will be used for preparing publications in leading international journals. The scientific data produced during the simulations will be stored via backup on hard disks, and will also be stored for longer terms (10+ years) on tape, using storage robots located at the Technische Informations- und Kommunikationsdienste (TIK). The infrastructure for data handling is secured by the University of Stuttgart.

2.5 Other information

None

2.6 Description of proposed investigations involving experiments involving humans, human materials or animals as well as dual use research of concern

Not applicable

2.7 Information on scientific and financial involvement of international cooperation partners

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4 Requested modules/funds

4.1 Basic Module

4.1.1 Funding for Staff

Research Staff

We apply for one Ph.D. position in the first three-year funding period (TV-L E13, 75 %).

This amounts according to ELAN to **€ 145,125**.

The student would be working full time on the proposed research project. His theoretical work will be supervised by Dr. Holm. The development of the necessary algorithms, the LB multiphase and EK parts will be supervised by Dr. Jain.

Student Assistants

Total: €18,000. We would like to hire two undergraduate students, each for 6 months per year at 40 hours per month, at a cost of around € 6,000 per year. The reason for this request is that there are many small, yet meaningful simulation jobs to do that do not require the full attention of a Ph.D. student. This would also allow us to follow the declared policy of the DFG to involve undergraduate students in research projects already early in their studies. In previous projects we had several scientific publications that had originated from work of our undergraduates.

4.1.2 Direct Project Costs

4.1.2.1 Equipment up to 10,000 Euro, Software and Consumables

Total: € 9,400 (the total amount for the first year). We apply for buying two GPU cards of the type Nvidia Quadro P6000 already in the first year of the funding period. These cost currently 4,690 € per piece. These are perfectly suited for our software ESPResSo, since several of our algorithms, and specifically the LB-EK part are optimised for GPU computations. Although we have a computing cluster available (resources see later), this machine is already 3 years old, and is in heavy usage for other projects. Installing two powerful GPUs of the modern architecture would allow us to speed up the computational aspects of our project largely.

4.1.2.2 Travel Expenses

Total: €15,000 (€5,000 per year). This amount is based on yearly travel of the two PIs plus the Ph.D. student. These travels include attending 1-2 national meetings (€500 each), and visiting the SPP events like the SPP summer school, SPP conference, or research stays at collaborating groups; also €500 each), and one international meeting (e.g.; ICMMES, Particles and/or DSFD each €1,500).

4.1.2.3 Visiting researchers

none

4.1.2.4 Expenses for Laboratory Animals

Not applicable

4.1.2.5 Other Costs

Not applicable

4.1.2.6 Project-related publication expenses

Total: €2,250 (€750 per year). This amount is based on the occasional costs for color printing, non-green-route open access, and cover images.

4.1.3 Instrumentation

None

5 Project requirements

5.1 Employment status information

Prof. Dr. Holm, Christian: W3 Professor, tenured

Dr.-Ing. Jain, Kartik: Scientist, fixed-term, contract ends July 2022.

5.2 First-time proposal data

Not applicable

5.3 Composition of the project group

- Holm, Christian; Prof. Dr.; Director of the Institute for Computational Physics (ICP); Universität Stuttgart, permanent, university funds
- Jain, Kartik; Dr.-Ing.; Senior Scientist at the Institute for Computational Physics (ICP); Universität Stuttgart, fixed term, currently third party funds
- Patzelt, Henriette; - ; Secretary; permanent, university funds
- Huber, Frank; - ; IT Administrator; permanent, university funds
- Tischler, Ingo; Ph.D. student, catalytic reactions, usage of the LB-EK, fixed term, third party funds
- Kuron, Michael; Ph.D. student, active particles, fixed term, third party funds

All other members of the ICP group work on non-project related scientific topics, notably on theory and simulations of charged soft matter systems, in particular simulations of polyelectrolytes, charged colloids, networks, and magnetic fluids and gels, but we have no other projects related to wetting. The standard simulation package used at the ICP is ESPResSo (www.espressomd.com), which was developed in the group of PI Holm. This package is publicly available under the GPL license and still actively extended by several members of the ICP and through external contributions by other users worldwide.

5.4 Cooperation with other researchers

5.4.1 Researchers with whom we have agreed to cooperate on this project

We see our contributions to the goals of the SPP 2171 in the following way: we are a computationally physics group that will develop a MD/LB-EK coupling scheme that can bridge the molecular scale to the continuum scale. In such a way, we can make contact to more mathematical theories working on the continuum scale, but at the same time we can also include molecular details into our models to see their influence onto the macroscopic scale. The understanding of experimental results will always rest on both scales, since normally they can easily modify interactions on the molecular scale, but the measurements are often performed on the macroscopic scale, and that is also the relevant scale for many envisioned applications.

During the first funding period of the SPP 2171, we plan to work intensively with several scientists that have submitted proposals to this SPP on the following topics:

- With the **Harting** group from the Forschungszentrum Jülich GmbH, Helmholtz-Institut Erlangen-Nürnberg, we plan to exchange knowledge and experience on the development and application of multiphase LB solvers for charged fluids.
- The group of **Huber** in Hamburg is providing us with data on the static droplet shape as function of electric field and electrolyte concentration, as well as on the droplet dynamics and on the electrokinetics (electrical capacity, ion kinetics) as well as on the nanopore imbibition kinetics. All these observables can also be measured within our simulations. Since the geometric response of the substrate to the fluid can be ignored as a first approximation, and we can simulate both, the porous and the non-porous surface, this system is very valuable in fine-tuning the input parameters from our LB-EK model. Our parametric studies can give a more microscopic and physical insight into the experimental results, since we have additional information about the space and time dependent variations of the ion concentrations and the electrical potential everywhere in the drop.

- The group of **Prof. Frank Müller** in Jena is planning to conduct experiments to study the dynamic wetting behavior of nano-structured ferroelectric polymers. They have agreed to provide us with experimental data, and we will use the results of our LB-EK model to study the behavior of their synthesised surfaces as a function of solvent pH and under an applied electric field. The data provided by them will also help us to validate our model.
- With the project of **Müller/Tanaka** we will compare the results on stimuli-responsive brushes and exchange ideas on novel simulation algorithms and coarse-graining strategies.
- The research group of **Monkman** from Regensburg will provide us with electric field (EF) data for our simulation studies. This group is equipped with means of generating and measuring magnetic and electric fields under various static and dynamic mechanical conditions. The EF data will thus consist of electric field magnitudes for various surfaces –, both AC and DC.
- **Dr. Vivek Pachauri** from the RWTH Aachen University proposes to study corrugation of graphene surfaces in a way that they have localised areas with hydrophobic and hydrophilic patches, which they will adjust by ionisation. They will then study the polymer's response to pH on these corrugated surfaces, which complements well with our simulation set-up. They will provide us with experimental data and we in return give them computational insights in this aspect.
- With the group of **Rühe** from Freiburg we have agreed to try to model also brush and gel coated surfaces with our two-phase LB-EK. We would try to incorporate the temperature stimulus into effective monomer interaction to mimic the change in polarity of their surfaces. The Rühe group will generate layers where the interaction with an external stimulus (light, heat) will change the polarity/hydrophilicity of a micro- and nano-structured surface and study the wetting properties of the thus changing layers from an experimental point of view, which will be a perfect match to our simulations.
- With the **Schönecker** group from the TU Kaiserslautern, we will collaborate on the modeling aspects. They plan to use continuum approach to simulate electric field driven wetting using finite element calculations, which can serve as a cross-verification of the numerical methods involved since we can compare those to the results coming from the particle-coupled LB-EK methods.
- The project of **Sega** from the Forschungszentrum Jülich GmbH, Helmholtz-Institut Erlangen-Nürnberg will study the contact line dynamics of a droplet of water using atomistic simulations. His approach is complementary to ours, since we use a mesoscopic approach. We will compare our results with the atomistic simulations since they can validate our parametrisation or show possible deficiencies. We, on the other hand, could upscale his results to the macroscopic size.
- From the group **von Klitzing** from TU Darmstadt we will obtain experimental data on the wetting angles of drops of varying electrolyte concentration on surfaces made up polyelectrolyte multilayers and microgels. The multilayers can have a positive or negative top surface, which we can easily incorporate into our models. She plans also to observe moving contact lines by tilting the probes, yielding different applied forces. We will modify our surfaces according her experimental setup and compare our results on wetting angles and drop shapes. She can also study the effect of pH on the dynamics of wetting, and we can qualitatively compare our results.

5.4.2 Researchers with whom we have collaborated scientifically within the past three years

- Günter K. Auernhammer, Leibniz-Institut für Polymerforschung Dresden e.V., Hohe Str. 6, 01069 Dresden
- Markus Bier, Hochschule Würzburg-Schweinfurt
- Oleg Borisov, Institut des Sciences Analytiques et de Physico-Chimie pour l'Environnement et les Matériaux, Pau, France

- Per-Kristian Eide (Oslo University Hospital, Oslo, Norway)
- Joachim Gross, Institut für Technische Thermodynamik und Thermische Verfahrenstechnik ITT, University of Stuttgart
- Joost de Graaf, Utrecht University in the Institute for Theoretical Physics, Utrecht, The Netherlands
- Steffen Hardt, Institute for Nano- and Microfluidics, Technische Universität Darmstadt, Darmstadt
- Andreas Heuer, Institute of Physical Chemistry, University of Münster, Münster, Germany
- Jingfeng Jiang (Michigan Technological University, USA)
- Ulrich F. Keyser, Cavendish Laboratory, University of Cambridge, JJ Thomson Ave, Cambridge CB3 0HE, United Kingdom
- Svyatoslav Kondrat, Department of Complex Systems, Institute of Physical Chemistry, Polish Academy of Sciences, Kasprzaka 44/52, 01-224 Warsaw, Poland
- Erin Koos, Department of Chemical Engineering (CIT), University of Leuven, Celestijnenlaan 200f - box 2424, 3001 Leuven, Belgium
- Peter Košovan, Department of Physical and Macromolecular Chemistry, Faculty of Science, Charles University in Prague, Hlavova 8 128 00 Praha 2, Czech Republic
- Andreas Kronenburg, Institut für technische Verbrennung, Universität Stuttgart
- Arnold J.T.M. Mathijssen, The Rudolf Peierls Centre for Theoretical Physics, 1 Keble Road, Oxford, OX1 3NP, United Kingdom
- Vartan Kurtcuoglu (University of Zürich, Switzerland)
- Kent-André Mardal (University of Oslo, Norway)
- Miriam Mehl, Institute for Parallel and Distributed Systems, University of Stuttgart
- Bryn Martin (University of Idaho, Moscow, USA)
- Andreas M. Menzel, Institut für Theoretische Physik II: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, 40225 Düsseldorf, Germany
- Stefan Odenbach, Chair of Magnetofluidynamics, Measuring and Automation Technology, Institute of Fluid Mechanics, Technische Universität Dresden, 01069 Dresden, Germany
- Thomas Palberg, Institut für Physik, JG Universität Mainz, Staudingerweg 7, 55128 Mainz
- Dirk Pflüger, Institute for Parallel and Distributed Systems, University of Stuttgart
- Rudolf Podgornik, Department of Physics, Faculty for Mathematics and Physics, University of Ljubljana, Jadranska 19, 1000 Ljubljana
- Wilson C. K. Poon, SUPA, School of Physics and Astronomy, The University of Edinburgh, King's Buildings, Peter Guthrie Tait Road, Edinburgh, EH9 3FD, United Kingdom
- Daniel Potts, Department of Mathematics, Chemnitz University of Technology, Chemnitz, Germany
- Sabine Roller (University of Siegen, Germany)
- Mathieu Salanne, Sorbonne Universités, UPMC Univ. Paris 06, CNRS, Laboratoire PHENIX, F-75005 Paris, France
- Pedro A. Sánchez, University of Vienna, Sensengasse 8/9, 1090 Wien, Austria
- Lars V. Schäfer, Center for Theoretical Chemistry, Ruhr-University Bochum, Germany
- Annette M. Schmidt, Universität zu Köln, Department Chemie, Institut für Physikalische Chemie, Köln
- Marcello Sega, University of Vienna, Sensengasse 8/9, 1090 Wien, Austria
- Sela Samin, Utrecht University, Utrecht, The Netherlands
- Marcello Sega, Department of Computational Biological Chemistry, University of Vienna
- Ulf Schiller, Department of Material Science and Engineering, Clemson University
- Tyler N. Shendruk, The Rudolf Peierls Centre for Theoretical Physics, 1 Keble Road, Oxford, OX1 3NP, United Kingdom
- Patrice Simon, UPS - CIRIMAT-LCMIE, 118 route de Narbonne, Toulouse, Frankreich
- Jens Smiatek, Institute of Physical Chemistry, University of Münster, Münster, Germany
- Charles Strother (University of Wisconsin, Madison, USA)
- Regine von Klitzing, Stranski-Laboratorium, Institut für Chemie, Technische Universität Berlin, D-10623 Berlin, Germany

- Manfred Wilhelm, Karlsruhe Institute of Technology (KIT), Institute for Chemical Technology and Polymer Chemistry
- Z. Xu (Shanghai Jiao Tong University, China)

5.5 Scientific equipment

For the proposed project we require computing power in large amounts and specifically on graphics Processing Units (GPUs). Locally at the Institute for Computational Physics (ICP), we can make use of around 50 computers that are all equipped with Nvidia Fermi, Kepler, and Maxwell GPUs and powerful Intel Sandy Bridge and Haswell Central Processing Units (CPUs), as well as a 47-node compute cluster with Nvidia Kepler GPUs, coupled to Intel Haswell CPUs. All cluster nodes are connected over a 4x Quadruple Data Rate (QDR) Infiniband network. We will, however, require to purchase a two new GPUs as the already available nodes are in heavy use by the ICP members in other projects.

If additional computing time is required, we will submit a “Rechenzeitanträge” to the High Performance Computing Center in Stuttgart (HLRS) and other computing resources (e.g., *bwUniCluster*).

5.6 Project-relevant cooperation with commercial enterprises

None

5.7 Project-relevant participation in commercial enterprises

None

6 Additional information

This proposal has not been submitted to any other third-party grant organisation.