In- and out-of-equilibrium simulations of polymeric molecules at the monomer if and the coarse-grained level

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Ultrasoft polymeric macromolecules show bounded effective interactions, which arise as the internal degrees of freedom of the molecule are averaged out. This means that they can fully overlap their centres of mass without violation of the excluded volume at the monomer level. Under certain conditions, these systems form cluster crystals: crystals whose lattice sites are occupied by clusters of partially or fully overlapping particles. Even though the macromolecules within a cluster repel each other, the cluster as a whole is stabilised by the repulsion of the neighbouring clusters. This counter-intuitive, but meanwhile well understood, behaviour leads to a novel class of materials which shows remarkable properties, as for example a density independent lattice constant or hopping mechanisms, where particles diffuse hopping from one cluster to another.

In this thesis we study certain dynamic properties of these materials on the coarse-grained level: firstly we examine the effect of hydrodynamic interactions (due to the presence of a solvent) on the diffusion and hopping mechanism in pure and binary cluster crystals. Secondly we study the response of a pure cluster crystal to an external compression, paying special attention to the mechanisms through which the crystal can accommodate a volume reduction while keeping the spacing of the lattice unchanged. We also present an in silico design of a polymer amphiphilic chain, whose effective interaction shows the necessary properties for the formation of stable cluster crystals. The amphiphilic chains are composed by a solvophobic backbone decorated by solvophilic side groups. We perform monomer resolved simulations of a bulk crystal of these chains and verify the stability of the system. Finally we focus our attention on the computation of effective interactions of polymeric macromolecules. We make use of a method, which allows for the computation of the effective potentials at finite density leading to a more reliable description of the behaviour of the system