Editorial: Multiscale simulation methods for soft matter systems

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Editorial

Editorial: Multiscale simulation methods for soft matter systems

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It is now widely accepted that multiscale modelling is the key to success in computational materials science. It is simply not possible, from a computational point of view, to study a material in full quantum mechanical detail on the length and time scales that are relevant for practical applications (millimetres or more, seconds or more). Simulations of materials therefore typically rely on a hierarchy of models that describe structural properties of materials and dynamicals processes on different scales at different levels of representation. The underlying idea is that these models can be simulated more efficiently since uninteresting fine-grained details have been removed [1, 2].

A central element of multiscale modelling is coarse-graining, i.e., replacing the high-dimensional phase space of the original, microscopic system by a highly reduced set of representative degrees of freedom or collective variables. The concept of coarse-graining is very old. Famous examples include the description of planets as point particles, the approximate description of particle dispersions by Brownian motion, the derivation of classical force fields from electronic structure calculations, or the transition from microscopic liquid models to hydrodynamic theories.

Despite these apparent successes, establishing a generally applicable and at the same time practically useful methodology of coarse-graining and multiscale modelling remains a formidable task. This becomes clear when inspecting more closely the theoretical foundations of coarse-graining: the first and already difficult task in coarse-graining is to define suitable coarse-grained (CG) variables, i.e., collective variables that are able to capture the desired properties of a material in a CG model. Having identified these variables, one can then proceed with coarse-graining either in a static or dynamic sense. The formal task of static coarse-graining at equilibrium is to calculate partial partition functions in constrained ensembles with fixed CG variables, and use these to determine a free energy landscape as a function of the CG variables. In general, however, it is not possible to derive simple analytic descriptions for this free energy landscape, e.g., write it as sums of effective pair or low order multibody potentials. Coarse-graining dynamical properties is even more difficult. The task then consists in deriving dynamical equations for the CG variables, which, on average, reproduce their true dynamics in the underlying microscopic ensemble. Projection operator tools to derive such equations have been developed already in the 60s by Zwanzig, Mori, and others [3–5]. They result in complex integro-differential equations that, again, usually cannot be reduced to simple stochastic dynamic equations without substantial further approximations [6–8].

The coarse-graining problem is thus a classic example of the famous 'no free lunch theorem', which states that it is not possible to simplify a complex problem by just reformulating it [9]. Coarse-graining is traditionally most successful when the relevant CG variables are obvious, and when the relevant time and length scales are clearly separated by orders of magnitude. In all other cases, simplifications must be made, and multiscale modelling effectively turns into an *optimization problem*.

The collection of papers in this special issue reflects the scientific programme of the Collaborative Research Centre SFB TRR146: *Multiscale simulation methods for soft matter systems* in the Rhine Main area of Germany (Mainz and Darmstadt). It has the mission to develop and analyse tools for set-

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ting up and optimizing multiscale simulation methods in a systematic manner, with a solid mathematical background. The special issue features articles from scientists working in this network as well as from further invited contributors who are doing fundamental research at the forefront of research in the field.

The materials considered in this issue are mostly soft matter, a class of materials where multiscale modelling is a particularly hard problem. Soft material are omnipresent in life and technology, with examples ranging from plastic, rubber, (bio)membranes, but also complex fluids like oil, paint, blood, liquid crystals, and nonequilibrium materials such as active colloids. Soft materials by definition exhibit strong and slow responses to external stimuli, implying that characteristic energies are low (of the same order than the thermal energy at room temperature), and entropy is as important as energy. They usually consist of large molecules or other nano-micrometre size constituents, and their behavior is governed by an intricate interplay of intermingled processes on many scales. Complex fluids made of soft materials typically exhibit structure on more than one characteristic length scale. Therefore, soft materials are suitable testbeds for developing and testing novel multiscale modelling approaches.

The articles in the present collection address several challenges that we consider to be particularly pressing in the field of multiscale modelling.

The first challenge is *structural coarse-graining*, where well-known issues are *representability* and *transferability*. The representability issue states that CG models which were designed to reproduce certain equilibrium features of a system, e.g., the local structure, often fail to reproduce other features, e.g., thermodynamic properties. The transferability issue describes the problem that CG models which were optimized for one state point, e.g., one composition, may not be suitable for describing the same system at another state point, i.e., another composition. Both issues are addressed, among other, by Szukalo and Noid [10], who propose a novel 'dual-potential' approach for analyzing CG configurations. By separating entropic and energetic contributions to the CG potential, they can predict energetic characteristics of systems more accurately from CG simulations, and they can extrapolate CG potentials from one state point to another. An option to enhance the transferability of CG potentials is to use multibody and/or density dependent potential. This is explored in two articles in this issue, one by Baul and Dzubiella [11] who derive density dependent CG potentials for systems of responsive colloids, and one by Berressem *et al* [12] who compare the performance of multibody potentials and density dependent potentials in simulations of homopolymer films.

A particularly intriguing representability issue arises in CG polymer models based on soft potentials: such CG models may largely overestimate the probability of self-entanglements (knots) of chains. [13] This is analyzed by Wu *et al* [14] for two popular classes of soft-core CG models, namely dissipative-particle dynamics and density functional based models. On the other hand, Tubiana *et al* [15] show that it is possible to restore fine-grained polymer configurations with the correct knot spectrum from configurations generated with soft-core CG models.

Structural coarse-graining also has an impact on the *dynamical properties* of a CG model, even when applied naively in a Hamiltonian framework. Coarse-graining free energy landscapes makes them smoother, which results in a speedup of dynamical processes. Moreover, different time scales move closer to each other—the telescope effect [16]. From a computational point of view, both effects have their advantages. However, in studies of kinetic pathways, it is important that at least the order of time scales remains the same. Kloth *et al* [17] and Rudzinski *et al* [18] have examined this for ionic liquids and found that accurate structural coarse-graining also improves the dynamic properties of CG models in regular molecular dynamics. Li *et al* [20] propose a method to adjust relative time scales in extremely CG Brownian polymer models by adjusting internal friction parameters. Bockius *et al* [19] introduce a novel, systematic, and mathematically well-founded approach to constructing non-Markovian CG models (with memory) from fine-grained trajectories.

Establishing the connection between *particle-based and continuum models* is a third challenge in multiscale modelling, which needs to be tackled if one wants to span a wide range of scales. Spiller *et al* [21] present a systematic derivation of a continuum model for viscoelastic phase separation from a well-defined molecular model. Brunk *et al* [22] analyze the properties of this model and related ones from a mathematical point of view and present results on the existence of weak solutions, conditional uniqueness, and stability.

The final challenge is to develop *multiresolution* approaches that combine different levels of resolution at a time, for studies of heterogeneous systems or systems far from equilibrium, Deußen *et al* [23] consider suspensions of active self-propelled particles and show how to use state-of-the-art discontinuous Galerkin methods to study the collective behavior of swimmers with arbitrary stress boundary conditions. Baptista *et al* [24] reconsider the established Hamiltonian adaptive resolution simulation (AdResS) method [25, 26] and establish a connection to density functional theory. Oestereich *et al* [27] show how AdResS can be used to study a classical non-equilibrium problem, the forced folding and unfolding of molecular complexes. In complex real-life applications, it is often advisable to incorporate experimental information in the construction of CG models. Giunta *et al* [28] demonstrate how this can be done in the case of a polymer nanocomposite, polyisoprene filled with carbon black.

The articles in this special issue illustrate the progress that has been made in the field of multiscale modelling of soft matter in recent years, as well as the many remaining challenges and open questions. They also demonstrate the potential of multiscale simulations for obtaining insights into complex systems and processes, such as nanoscale-segregated ionic liquids [17], high performance rubber materials [28], emergent properties of active or responsive colloids [11, 23], or controlled nanoparticle growth [29, 30].

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Data availability statement

No new data were created or analysed in this study.

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