Machine Learning Polymer Force-Fields

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Background. Organic electronics require macromolecules which have conjugated backbones to facilitate the delocalization of electron orbitals and the transport of charges along the backbone. Conducting polymers (CPs) offer inexpensive alternatives to inorganics but control over the semi-crystalline structures formed and the link between molecular level morphology and material properties remains poorly understood. One of the most widely used polymers at present is poly(3-hexylthiophène) (P3HT) due to its conductive properties when doped and ease of manufacture at industrial scales. This makes it attractive for use in organic devices such as Solar Cells or Thermoelectrics. It is also of significant interest to experimental groups (SYCOMMOR) working within the ICS [1] and the wider Cronenbourg community. Molecular simulations could provide a testing ground where polymer crystals, representative of real experiments, can be grown and structure-property relationships understood with precision. The Theory and Simulation of Polymers Group at ICS has a strong track record in the study of structure formation and mechanical properties of polymers.

Accessing length scales typical of such crystals (>100nm) requires very large simulation boxes with large numbers of atoms and multiple potentials. A coarse-grained (CG) model is therefore required, which contains enough physical information to reproduce the correct structure whilst rendering simulations computationally tractable. Thus far studies which have attempted to grow P3HT crystals have fallen short of reproducing crystalline phases, instead only mesophases, with poor long ranged ordering have been seen [2,3]. A fully representative CG model, of CPs such as P3HT, is therefore necessary and the aim of this project is to build one by extending the united monomer (UM) model of polyethylene, recently used to grow the largest polyethylene (PE) single in molecular simulations to date [4,5].

PhD project. A coarse-grained model with explicit side chains of P3HT usable in molecular dynamics shall be developed. Intermolecular interactions between P3HT backbones will be extracted using the traditional Boltzmann Inversion method, from small all-atomistic simulations of P3HT and the CG force-field subsequently tuned using simple Machine Learning algorithms. Once a working force-field has been built, crystallization studies of monodisperse P3HT chains will be performed, using different crystallization protocols, to test the force-fields common attributes with real experimental atomic force microscopy (AFM) data from Dr Martin Brinkmann in the SYCOMMOR group at the ICS [1]. It will be possible to connect morphological features such as crystallinity and tie-chain content to real experimental data in this way. The PhD thesis will be co-supervised by William Fall from the LPS in Orsay.

Profile. The PhD project involves extensive computer simulations and the training of ML algorithms as well as data analysis based on advanced theoretical concepts. The candidate must have a disposition for numerical work and theory. A good background in statistical physics and in programming is required. Experience with either LAMMPS/Keras or PyTorch is desirable.

[1] Guchait, S., Zhong, Y., & Brinkmann, M. (2023). *Macromolecules*, *56*, 6733-6757.
[2] Greco, C., Melnyk, A., Kremer, K., Andrienko, D., & Daoulas, K. C. (2019). *Macromolecules*, *52*, 968-981.
[3] Wood, E. L., Greco, C., Ivanov, D. A., Kremer, K., & Daoulas, K. C. (2022). *The Journal of Physical Chemistry B*, *126*, 2285
[4] Fall, W. S., Baschnagel, J., Lhost, O., & Meyer, H. (2022). *Macromolecules*, *55*, 8438-8450.
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