Master 2: International Centre for Fundamental Physics

Internship Proposal

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Thesis possibility after internship: YES
Funding: Not Currently

Molecular Simulations of Chiral Self-Assembly in Binary CNC-Polymer Mixtures

Cellulose is the most abundant polymeric raw material on the planet and is found almost exclusively in the cell-walls of plants. It is environmentally friendly and sustainable to produce, on industrial scales and can be processed into nano-structured materials. This makes it one of the most prominent 'green' materials of modern times to be used in bio-based functional materials [1].

The polymer itself is inherently chiral, with a preferred helical twist and is known to form bundles known as cellulose nanocrystals (CNCs). These CNCs exhibit rod-like liquid crystalline (LC) behaviour and spontaneously order into cholesteric phases with left-handed macroscopic chirality. Such a transfer of chirality across length-scales is an intriguing natural phenomenon but connecting small-scale molecular details to large-scale structures remains a real challenge [2]. In fact, the precise nature of the twist geometry in CNCs is still a matter of debate due to contrary experimental observations [3]. One of the aims of the project is to provide molecular level insights into the large-scale LC behaviour of CNCs and to understand the effect of depletion on the transfer of chirality across length-scales. We will also explore the morphology of mesoscale droplets formed by CNCs and non-adsorbing polymers. The presence of the polymers imparts strong depletion attractions between the CNCs which may potentially lead to droplets with reconfigurable shape such as membranes and twisted ribbons [4], see Fig. 1.

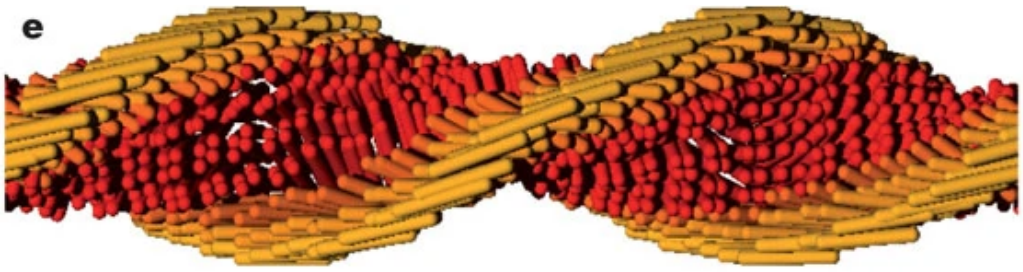

Figure 1- Schematic structure of twisted ribbons taken from [4].

Hybrid coarse-grained molecular dynamics (CGMD) simulations, of binary mixtures of polymers and chiral CNCs, will be performed using the Jean-Zay supercomputer in Orsay, using a combination of flexible and rigid body dynamics in LAMMPS. Explicit chirality will be incorporated by tuning the twist of the point-mesh of the rods, see above for an example.

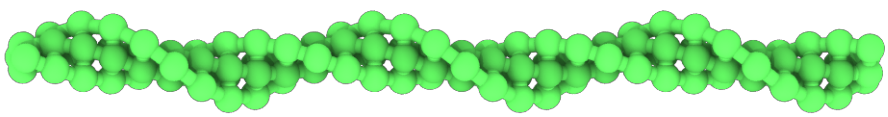

Figure 2 - A coarse-grained representation of a twisted CNC.

Applicants should have a sound understanding of classical statistical mechanics and thermodynamics and an appetite for theoretical/numerical computations. Some experience with Linux, Python, C/C++ and LAMMPS is desirable but non-essential. Candidates can expect to learn the basics of MD simulations and the fundamental importance of chirality in nature. Early application is encouraged due to the administrative procedure required to access GENCI resources and the possibility of additional background checks. Some interaction with the recently launched international INTEGRATE consortium, funded by EIC Pathfinder, may be required. https://www.integrate-pathfinder.com

[1] Trache, D., Tarchoun, A. F., Derradji, M., Hamidon, T. S., Masruchin, N., Brosse, N., & Hussin, M. H. (2020). Nanocellulose: from fundamentals to advanced applications. *Frontiers in Chemistry*, *8*, 392.
[2] Parton, T. G., Parker, R. M., van de Kerkhof, G. T., Narkevicius, A., Haataja, J. S., Frka-Petesic, B., & Vignolini, S. (2022). Chiral self-assembly of cellulose nanocrystals is driven by crystallite bundles. *Nature Communications*, *13*(1), 2657.
[3] Ogawa, Y. (2019). Electron microdiffraction reveals the nanoscale twist geometry of cellulose nanocrystals. *Nanoscale*, *11*(45), 21767-21774.
[4] Gibaud T. et al. (2012). Reconfigurable self-assembly through chiral control of interfacial tension. *Nature 481*, 348-351.