Dissipative self-assembly of C_3-symmetric discotic molecules

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Introduction: Supramolecular chemistry has allowed us to produce very intricate ***structures*** that rival the architectural complexity of biological ones, by using self-assembly of synthetic artificial molecules.[1] So far, however, self-assemble structures that have ***functions*** that are as complex as those found in living systems are lacking. The main reason for this is that so far we have studied systems in thermodynamic equilibrium (State #1, Fig. 1a) or in kinetically trapped / metastable states (State #2, Fig. 1a), all of which are formed during a dissipative (i.e., an energy consuming) process, but the resulting structures themselves are non-dissipative. Living systems, on the other hand, are in a dissipative nonequilibrium state (State #3, Fig. 1a) and continuously consume energy to keep their ***structure and function*** at the expense of increasing the entropy of their surroundings.[2]

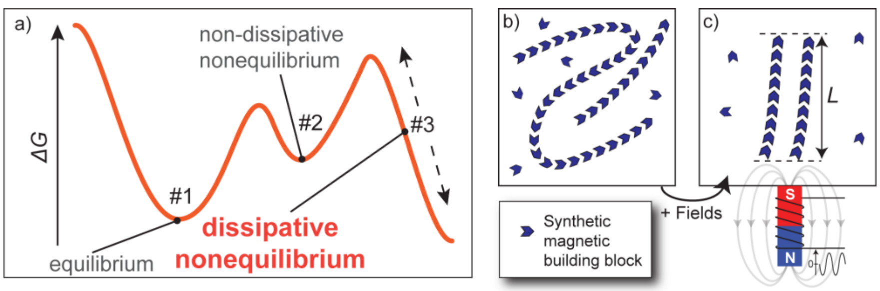

Fig. 1 | a) The three different thermodynamic states.[2] b) without an AC field the molecule will form an equilibrium supramolecular polymer. c) with an AC field, the structures with a length corresponding to the frequency of the field will selectively be disassembled. Only non-resonant structures of length *L* will remain.

Recently, we and others have investigated a range of C_3-symmetrical benzene-1,3,5-tricarboxamide discotic molecules that self-assemble into helical supramolecular polymers with a strongly cooperative mechanism.[3,4] We propose to use the same molecular design but form complexes with rare earth ions, leading to significant magnetic dipole-dipole interactions when the moments are in close proximity. *We propose to use oscillating fields to control the structure and dynamics of supramolecular fibers (Fig. 1b,c)*. This will lead to dissipative nonequilibrium supramolecular states (State #3, Fig. 1a), which have barely been explored.

Work plan: The PhD student to be recruited will work on the following aspects:

- Study equilibrium self-assembly of the discotic molecule for different rare earth ions (yr 1)
- Study equilibrium self-assembly in a constant magnetic field (yr 1)
- Build an AC magnetic field setup that will be incorporated into a light scattering setup (yr 2)
- Study dissipative self-assembly of discotic molecule in AC field setup (yr 2, 3)

Deliverables: *In this project, the PhD candidate will demonstrate how dissipation can be achieved in supramolecular assemblies using a magnetic field, and how this can lead to control over structure and dynamics. This project is at the frontier of supramolecular chemistry.*

References

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