LIGHT FROM WITHIN: ILLUMINATING THE COMPLEXITY OF CO-ASSEMBLY FROM THE INSIDE OUT

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The assembly of well-defined nanostructures from smaller (macro)molecular building blocks relies on a delicate balance between attractive and repulsive interactions between the basic building blocks. To avoid kinetic trapping and structural polymorphism, the balance between antagonistic and synergistic interactions must be carefully tailored. Controlling this complex process is challenging, but we can look to Nature for inspiration. As it turns out, biology often use a combination of molecular templating and allostery to control self-assembly processes, resulting in an unusual degree of fidelity of the final nanostructures formed.

In this contribution, I will discuss how templated assembly and allostery interplay in supramolecular co- assembly. I will demonstrate how the complex supramolecular reaction cascade that underlies templated assembly can be illuminated from within using semiconducting macromolecules as molecular sensors. I will first discuss the force-optical response of these sensor molecules and how they can be used to probe supramolecular forces in solution. I will then discuss two particular examples of complex co-assembly processes that we have illuminated using our molecular sensor toolbox: i) the electrostatic condensation transition during nanoscale charge complexation and ii) the reaction pathways of viral capsid assembly.

I will conclude my talk with an outlook for the use of molecular force sensors to probe a wide variety of complex soft matter problems, including targeted sensor binding to biological structures, probing contact forces between colloidal particles, and force-sensing at the single-molecule scale.