Intermolecular forces drive self-organization of molecules, which is ultimately the origin of most the physical and chemical phenomena in Nature\(^1\). Molecules able to interact themselves by non-covalent forces, as hydrogen bonding and/or hydrophobic attractions, usually form macrostructures in condensed phases (solid, solution). The properties of these aggregates depend of three main factors: the structural and chemical features of the molecules, the nature of intermolecular forces and the environment. The first two drive aggregation in solid state, while in solution the role of the solvent become determinant as it can induce a variety of structural effects on the aggregation behaviour of the solute\(^2\). In the case of chiral molecules, this property is transferred to the aggregates and supramolecular chirality appears.

Here we present our research on chiral molecules that self-organize in solution forming helical structures. We use VCD as the main chiroptical tool, but also supported by other chiroptical spectroscopies (ECD, ROA) and theoretical modelling. In our first steps, we studied the effect of modulating the environmental settings on the helices. Thus, helix handedness was proved highly and reversibly dependent on factors as pH or ionic strength in peptide-mimetic hydrogelators. We also observe how the initial conditions (concentration, temperature) were capable of controlling the helix structure of oligo-p-phenylene-based polymers towards kinetic or thermodynamics pathways. Besides, the structure of the helices can also be the consequence of direct solvent-solute interactions. In this way, we have demonstrated that an achiral solvent can act as a template for chiral organization of N-heterotriangulenes-based organogelators, thus showing the different levels of complexity of the hierarchical organization of supramolecular polymers.

But the solvent-helix interactions can be bidirectional. As a nice example, we recorded chiral signals which can be only assigned to the organization of the solvent molecules around helical aggregates of phenylglycine functionalized poly(phenylacetylene)s. The solvent molecules thus form a first solvation shell to which the helix chirality is transferred. The helices would act therefore as a template of the solvent molecules, and the chirality of this external helix would be fully controlled by the solute.