Biological materials are often used as inspiration in the design of new synthetic or bio-hybrid materials; however, the molecular toolbox provided by biological systems has been evolutionarily optimized to carry out the necessary functions of cells. The resulting inability to systematically modify fundamental properties such as polymer stiffness or the association strength of crosslinking proteins in experimentally available model systems hinders a meticulous examination of the connection between molecular parameter space and resulting properties of bulk assemblies. We circumvent these limitations using model systems based on synthetically produced building blocks such as DNA strands and peptides, which are programmable on the molecular scale.

In one example, micrometer-long nanotubes with tunable diameters and rigidity can be constructed from small sets of short, DNA oligonucleotides. By systematically varying the set of DNA strands of these synthetic, semiflexible filaments, their micron-scale persistence length ($L_p$) can be precisely tuned. In low-density entangled networks [1], this allows control of network elasticity ($G'$) over two orders of magnitude. While the scaling of $G'$ with respect to network density supported predominant models, the $L_p$ scaling, here measured experimentally for the first time, showed a linear dependence, which stands in contrast to the strongly sublinear behavior predicted by long-accepted theory. When assembled at high density in the presence of a molecular crowding agent [2], concurrent nucleation and growth processes lead to the formation of bundled, star-like and compact microparticles, whose morphology and phase behavior depend upon the nanotube persistence length.

Hybrid molecules consisting of DNA and peptides can also be used to control mechanics and morphology of biopolymer networks [3]. Synthetic constructs for crosslinking actin filaments are fabricated from DNA strands which have been conjugated to actin-binding peptides. These were shown to modulate bulk network elasticity and mesoscopic assembly in accordance with binding strength, concentration and size of the crosslinking construct. Furthermore, these were found to mimic certain non-canonical, time-dependent ageing behaviors of crosslinked biopolymer systems.