The transfer of chirality from the molecular scale to the mesoscale of self-assembled materials is common theme to a wide range of molecular systems, from cholesteric liquid-crystalline materials to structure and assembly of proteins. In this talk, I will discuss recent theoretical studies of two self-assembling polymer systems where the tendency to form a chiral, or "twisted", intermolecular order frustrates or competes with the ideal packing. In both cases, the chiral frustration gives rise to a much richer thermodynamic behavior than occurs in the achiral assemblies, pointing towards unusual mechanisms for controlling molecular organization across multiple length scales.

In the first example, I will discuss the frustration of rope-like, cohesive assemblies (bundles) formed from stiff and chiral molecular filaments, such as cytoskeletal or extra-cellular protein filaments. In these materials, intrinsic twist of the bundle makes perfect lateral packing impossible and stabilizes a rich spectrum of heterogeneous, defect-riddled ground states whose structure is intimately (and surprisingly) connected with classical Thomson problem of packing on spherical surfaces. In the second example, I will discuss how introducing chirality at the monomer scale has a profound impact on the now well-known phase diagram of block copolymer melts. In particular, I will explore the meso-scale transfer of chirality in a recently discovered chiral mesophase, the H*, or helical cylinder phase. I will discuss how the complex thermodynamics of chiral transfer in block copolymers melts derive from the subtle threading of cholesteric segment packing within chiral mesodomain.