The design and development of adaptive functional structures using artificially synthesized nano-constituents as building blocks is one of the most compelling current research areas. The typical approach is bottom-up assembly of nanoparticles (NPs), and while this has led to the successful creation of two- and three-dimensional super-lattices, these tend to simply exhibit the combined properties of the constituent NPs, instead of exhibiting a novel functionality arising from synergistic inter-particle interactions. The route we use involves utilizing soft materials such as liquid crystals as templates for NP assembly, which removes rigid constraints in the way the individual components can be combined and make novel architectures possible, such as non-planar structures. Additionally, it allows us to modulate the NP assembly in situ via external controls, such as temperature, mechanical strain and electromagnetic fields, which lead not just to greater versatility in functionality, but to the emergence of entirely novel behavior. Following this approach, we demonstrate “cluster assembly” of metallic, magnetic and semiconducting NPs that form structures with interesting new properties and diverse applications in magneto-optical sensing, photothermal therapy, and tissue engineering.