Graphene, a single layer of carbon atoms arranged in a hexagonal lattice, is probably the best known, and most extensively characterized two-dimensional material. However, this represents just one of a larger class of layered materials, in which weak interlayer-forces make it possible to mechanically isolate monolayers from the bulk. This weak interlayer bonding also makes it possible to interleave monolayers from different crystals together to form entirely new layered structures. Fabricating materials by the mechanical assembly of individual layers provides a new and unprecedented level of control in device engineering where crystals with wildly different properties can be mixed and match, virtually at will. In this talk I will describe some of the techniques we have developed to make this possible and highlight new quantum phases that emerge as a result. I will additionally discuss recent efforts where, by tuning the geometry of these heterostructures at the nanoscale, we realize the capability to induce and dynamically control novel electronic phases in these systems, in ways that are not possible in conventional materials.