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Supramolecular and covalent assembly of molecules at surfaces: chirality, complexity and diversity

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The nanoscale details of how complex molecular organisations and architectures are nucleated, controlled and propagated at surfaces have now begun to emerge from scanning probe microscopy, a powerful range of surface science techniques and periodic density functional theory. Organised molecular assemblies give rise to important functions such as molecular recognition, chirality, adaptive behaviour and confined motion that are important in functional interfaces and new materials. Furthermore, complexity in organisational behaviour is found to be induced by simple initiators such as the molecular interaction with the surface, chemical fields or from fluctuating populations of differing molecules. Finally, the transition towards robust, covalent assemblies will be demonstrated via on-surface synthesis of macromolecules using clean, generic connection strategies that utilise the C-H bond as a synthon. Scanning tunneling microscopy reveals that covalent macromolecular heterostructures, displaying diverse compositions, structures and topologies, are created with ease from seven distinct building blocks, including porphyrins, pentacene and perylene. By exploiting differences in C-H bond reactivity, controlled synthesis of specific products, such as block and cross-linked copolymers, can be attained. Further, the symmetry and geometry of the molecules and the surface can be exploited to determine the outcome of the covalent bond forming reactions. This strategy opens up the capability to generate libraries of multivariate macromolecules directly at a surface that, in conjunction with nanoscale probing techniques, could accelerate the discovery of functional interfaces.