Graphene, a single, atomically-thin layer of graphite, is the most researched material today. Since 2004 - when it was isolated for the first time, ground breaking experiments of fundamental science were followed by a cascade of demonstrations of potential usages of graphene in day-to-day applications. It was in this domain that my research lied: the central theme of my work, “Engineering the Wavefunction in Graphene Systems”, was to probe, engineer and harness the very special behaviour of electrons within the graphene sheet (the so-called “Dirac electrons”) and by doing so to lay down the foundations of completely new technologies. I studied the modulation of the wavefunction in bilayer and trilayer graphene systems originating from two underlying mechanisms: quantum interference phenomena (QIP) and quantum confinement. I also took a bottom-up approach to tailoring surface potential distributions at the atomic scale to influence/control electron behaviour, by utilising the interaction between graphene layers and nanostructured, atomically flat insulating ionic surfaces. Quantum interference phenomena were explored at bilayer-trilayer armchair interfaces in multilayer graphene with various stacking orders by using scanning tunnelling microscopy and with support from theoretical simulations. Effects of various types of edges, which terminate the stacks abruptly or appear at lateral interfaces within the multistack, were revealed and correlated with scattering mechanisms, while a taxonomy of interference patterns was established based on stacking order. The effect of extra sources of scattering was also studied to understand the origin of the well-known $(\sqrt{3}\times\sqrt{3})R30°$ superstructure in graphene systems, and a new explanation was proposed. The energy dependency of the $(\sqrt{3}\times\sqrt{3})R30°$ superstructure and its motifs was quantitatively explored in bilayer graphene. My results on quantum interference phenomena at lateral interfaces and in bulk within multi-stacked graphene systems can be envisaged as providing unique system-specific opportunities for wave-function engineering to be exploited in devices employing quantuminterference and its impact upon transport characteristics. Furthermore, graphene was overlaid on atomically flat ionic insulating surfaces to exploit the interaction between the two. It was demonstrated that the density of states of graphene was modified due to formation of superlattices and nanostructures from the underlying substrate.