"Over the past decades, continued improvement in transistors density, performance and energy efficiency have been sustained by technological innovations. This exponential development predicted by Moore in the 60s transformed the first crude computers from the 70s into the ubiquitous and high-performance sophisticated machines of nowadays. This trend is however coming to end as the size of transistors is reaching the quantum limit. In the last decade, two-dimensional (2D) crystals have emerged as a disruptive materials platform. Those layered crystals are excellent candidates for next-generation technologies due to their ultimate thinness and dangling-bonds-free surface. In addition, they offer better scaling than traditional 2D electron gas based on III-V compounds. In spite of the aforementioned advantages, some technological obstacles still hinder the industrial integration of 2D materials in electronics. The most important ones are the growth and the post-processing of wafer-scale high-quality crystals, robust passivation schemes and device-related aspects such as the contact resistance or doping.

In this thesis, first-principles simulations are performed on systems composed of graphene or transition metal dichalcogenides (TMDs). Their transport properties are computed using the OpenMX code based on the Green's functions formalism. First, metal-graphene interfaces are studied in the top-contacted geometry. Charge injection from metal to graphene is understood through the lens of a semi-empirical transport model so that the transport mechanisms impeding the contact resistance are identified. Afterwards, quantum transport is investigated inside the graphene channel. The objective is to benchmark the transport characteristics of polycrystalline samples resembling the ones produced by the industry. Finally, the operation of a TMDs-based atomically thin FET is investigated. Stain-induced current modulation is predicted through nano-scale quantum engineering, a mechanism not yet observed in TMDs-based lateral junctions.

The overall objective is to provide a deeper understanding of some degrading factors in the low-dimensional limit while proposing novel functionalities for 2D crystals architectures."