Beating the size limits of first-principles calculations in nanoscale systems

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The advances in the predictive power, speed and reliability of ab-initio methods has occurred in the last few decades at a very fast pace. Simultaneously, the computing power available through HPC facilities has continued growing exponentially. This combination has brought the paradigm of ab-initio simulations as an invaluable tool to understand and predict the behavior of matter at the nanoscale. First-principles electronic structure methods have advanced in their efficiency to the point where realistic simulations can now be done for systems with many hundreds of atoms [1]. Besides, these methods have been successfully extended to deal with nonequilibrium processes such as electronic transport [2]. Current-voltage characteristics for systems as large as several hundreds of atoms, as those shown in Figure 1, can be currently studied a the fully atomistic and first-principles level [3]. However, enormous challenges are still ahead of us, to be able to extend the range of practical applicability of these methods to the sizes and time scales which are relevant to most of the practical problems in nanotechnology. For instance, many of the processes and properties that make graphene an outstanding material for potential applications are still beyond the reach of these methods, due to the large length or time scales involved. One example is electronic transport, in which the scattering lengths involved are so large that straight first-principles transport calculations are not relevant, because they can still not reach the appropriate length scales. In this talk I will review work done in our group in using first-principles methods for the study of graphene (including electronic transport [3]), and efforts to extend these studies to reach larger length and time scales. In particular, I will describe an implementation of a hybrid QM/MM approach [4] and its application to the immobilization of proteins on graphite surfaces decorated with gold nanoclusters. I will also illustrate the use of first-principles calculations to obtain tight-binding parameters that can be used to compute the transport properties of large samples of chemically modified graphene [5-7].

References

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Figure 1. Pictorial representation of a device consisting in an array of carbon nanotubes linking two semi-infinite graphene sheets. The electronic properties of these arrays were studied using first-principles calculations in Reference [3]