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Review

Ab initio quantum transport calculations using plane waves

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ABSTRACT

We present an *ab initio* method to calculate elastic quantum transport at the nanoscale. The method is based on a combination of density functional theory using plane wave nonlocal pseudopotentials and the use of auxiliary periodic boundary conditions to obtain the scattering states. The method can be applied to any applied bias voltage and the charge density and potential profile can either be calculated self-consistently, or using an approximated self-consistent field (SCF) approach. Based on the scattering states one can straightforwardly calculate the transmission coefficients and the corresponding electronic current. The overall scheme allows us to obtain accurate and numerically stable solutions for the elastic transport, with a computational time similar to that of a ground state calculation. This method is particularly suitable for calculations of tunneling currents through vacuum, that some of the nonequilibrium Greens function (NEGF) approaches based on atomic basis sets might have difficulty to deal with. Several examples are provided using this method from electron tunneling, to molecular electronics, to electronic devices: (i) On a Au nanojunction, the tunneling current dependence on the electrode–electrode distance is investigated. (ii) The tunneling through field emission resonances (FERs) is studied via an accurate description of the surface vacuum states. (iii) Based on quantum transport calculations, we have designed a molecular conformational switch, which can turn on and off a molecular junction by

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applying a perpendicular electric field. (iv) Finally, we have used the method to simulate tunnel field-effect transistors (TFETs) based on two-dimensional transition-metal dichalcogenides (TMDCs), where we have studied the performance and scaling limits of such nanodevices and proposed atomic doping to enhance the transistor performance.

Contents

1. Introduction

The field of electronic transport through nanometer-scale systems, such as molecular junctions or atomic wires, has been an extremely active area during the last decades. Although a great number of advancements have been made both from the fundamental and the application point of view [1–5], there is still much to learn before nanoelectronics and, in particular, molecular electronics can replace the silicon based technology. The effect that the development of a post-silicon era might have on our daily lives, together with the existence of a number of extremely interesting scientific and technological open questions, indicates a bright future of this field [3,4].

The experimental investigation of electron transport across nanosystems was launched with the advent of the scanning tunneling microscope (STM) [6], which enables the measurement of current across a single molecule under the application of an external bias voltage. A few years later, break-junction techniques emerged as alternative ways to measure the transport across molecular bridges [7,8]. All these techniques have proven to be extremely powerful and, consequently, are still the prime tools for the measurement of transport properties across nanosystems. Recent experimental advances include the demonstration of various interesting effects, such as conductance switching, rectification, and interference effects [9–15].

Along with the huge amount of experimental activity, an equally major effort has been devoted to the development of computational methods appropriate for the theoretical investigation of electron transport at nanoscale. Due to the difficulty of experimentally probing the detailed atomic structure of the nanosystems, theoretical modeling plays a key role in the understanding and interpretation of experimental observations [1,5]. After the first semiempirical studies [16–18], *ab initio* simulations based on first-principles have been extensively developed. In particular, given the high sensitivity of conductance to atomic-structure details, density functional theory (DFT) has become an essential