COMPUTATIONAL NANOELECTRONICS WITH NON-EQUILIBRIUM GREEN'S FUNCTION METHOD

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Abstract: The progress in Nanotechnology has also made its way in the computational tools used in this scientific endeavor. The non-equilibrium Green's function (NEGF) method is such a technique that has become a standard computational tool in Nanoelectronics. In this communication we present the NEGF method and its applications to electron transport in a multi-layered structure (i. e., double barrier tunneling device) and in a molecule connected to metallic contacts.

Keywords: nano electronics, nanotechnology, semiconductors, non-equilibrium Green's function, Green's function, molecular electronics.

1. INTRODUCTION

The continuous downscaling of electronic devices has reached the deep nanometer-size regime, where quantum phenomena and atomistic models are needed in order to accurately model the electronic transport in these devices. According to the International Roadmap for Semiconductors (ITRS) in 2018 the critical length for transistors will need to be less than 10 nm to keep up with the Moore's law [1]. In this context an electron device is a quantum system coupled with at least two leads that play the role of reservoirs. Thus, the theory of charge transport can be viewed as an open system which can exchange locally conserved particles with environment. Moreover, the working conditions of these devices will be far-from-equilibrium meaning that the device coupled to at least two different particle reservoirs is brought and maintained in a non-equilibrium state.

To specify our discussion, by an open quantum system it is meant the device that fills up a finite region of space and exchanges particles on the boundary with the reservoirs (Fig. 1). Thus, the theoretical approach in treating the open systems is focused on the definition of boundary conditions defined in transport theory. One way to deal with the boundary conditions (BC) is to avoid the issue by treating spatially uniform systems, hence all spatial derivatives as well as the need to specify the BC vanish. This kind of treatment has been applied to Boltzmann equations under uniform fields [2, 3]. Periodic BC may be another approach [4], but it preserves the hermiticity of all operators, case that is characteristic to closed systems. Still the approach is appropriate for linear-response theory [5] but is inappropriate for studying systems far-from-equilibrium.

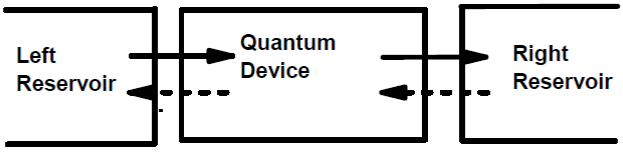

Fig.1. A generic quantum device coupled to the left and right leads associated as particle reservoirs.

The approach which really considers the open nature of transporting systems is due to Landauer [6,7], Fisher and Lee [8], and Büttiker [9,10]. The treatment is dissipationless with the device coupled to two or more particle reservoirs. The main quantity of interest is the conductance that has a quantum mechanical expression as a function of transmission coefficients. The BCs are given by imposing scattering states in the reservoirs, which are assumed to have defined thermal states. Low temperature experiments like the Aharonov-Bohm oscillations, universal conductance fluctuations, or quantized conductance through constrictions can be successfully explained by invoking the Landauer approach [11]. However, the Landauer formalism works just in linear-response regime (low temperature and low bias-voltage) which is close to thermal equilibrium.

A rather complete approach to quantum transport theory is based on the Green's-function (GF) techniques used in many-body physics. The non-equilibrium Green's-function (NEGF) theory was first formulated by Kadanoff and Baym [12] and then in a slightly different form by Keldysh [13] and later by Langreth [14]. The NEGF theory, defined by the correlation functions of single-particle creation and annihilation, describes completely the (quantum) kinetics of the systems including dissipative processes and memory (i. e., non-Markovian) effects. The appeal of the NEGF theory resides in its possibility of going beyond the single-particle picture to include electron-electron and electron-phonon interactions. For non-interacting electrons and without inelastic scattering given by the electron-phonon interactions the NEGF and Landauer formalisms are equivalent [15].

In this communication we apply the NEGF formalism both at semi-empirical level expressed in the tight-binding approximation and in an ab-initio setting given by the density functional theory (DFT) method. The tight-binding formulation is implemented in the open source software NEMO [16] and it is applied for studying the electron transport in a double-barrier semiconductor heterostructure. The ab-initio implementation is at the level of DFT using the SIESTA [17] and TranSIESTA [18] codes augmented with the inelastic transport due to phonon scattering [19]. With this ab-initio method we calculated the inelastic electron tunneling spectroscopy (IETS) signal of a molecule placed between two metallic electrodes. The paper has the following structure. In the next section the NEGF formalism is briefly reviewed. In the Section 3 and 4 two examples are presented. We end this paper with the conclusions.

2. BRIEF REVIEW OF NEGF THEORY

The term Green's function has several meanings. First, it is a powerful method for solving inhomogeneous differential equations in which the Green's functions (GFs) obey the differential equations with a singular inhomogenity that is a delta-function [20]. In quantum mechanics of non-interacting particles the Green's function is related to the inverse of the hamiltonian [21]. In quantum many-body physics the Green's functions are the correlation functions of fields or of creation and annihilation operators [20, 21]. Thus, the many body definition coincides with the one from quantum mechanics of non-interacting particles only for two-point Green's functions of non-interacting systems [20]. The Green's function technique turns out to be very a powerful tool for evaluating properties of many-body systems both at zero temperature (T=0) and in thermal equilibrium [20, 21] but also in the non-equilibrium situations [22].

Structurally equilibrium GF and NEGF are similar. Like in the equilibrium case, the NEGFs are defined as the correlation functions of single-particle creation and annihilation operators describing, in principles, completely the system. We encounter four GF in the NEGF theory. Thus, the GF $G^{<}$ (G less) provides the density of electrons while its conjugate $G^{>}$ provides the density of holes, G^{r} (G retarded) describes future time evolution while G^{a} (G advanced) gives the backward time evolution. These functions obey corresponding Dyson equations, which are also amenable to a perturbative approach. Instead of time-ordered GF, in the NEGF case the basic quantity is the path ordered GF which can be decomposed into expressions depending on those four fundamental Green's functions mentioned above [14, 22]. Let's consider the hamiltonian of the system can be written as a sum of two terms

$$H = H_0 + H'(t), \tag{1}$$

where H_0 is the solvable part and $H'(t)$ is the perturbation containing the external action as well as all many-body interactions of the system. The path ordered Green's function in the interaction picture is defined as

$$G^{P}_{\alpha,\alpha'}(t,t') = \frac{-i}{\hbar}\left\langle P e^{\frac{-i}{\hbar}\int_C ds H_I'(s)} c_{I\alpha}(t) c^{+}_{I\alpha'}(t') \right\rangle, \tag{2}$$

where P is the path ordering operator, C is the Keldysh contour, $H_I'(s)$ is the perturbing hamiltonian, and $c^{+}_{I\alpha'}(t')$ and $c_{I\alpha}(t)$ are creation and annihilation operators in the interaction picture. The brackets ⟨…..⟩ indicate the non-equilibrium average. The path ordered GF obeys the Dyson equation:

$$G^{P}(t,t') = g^{p}(t,t') + \int_C ds \int_C ds' \left[g^{P}(t,s) \Sigma^{P}(s,s') g^{p}(s',t') \right]. \tag{3}$$

All the real-time self-energies that are needed, $\Sigma^{<}$, $\Sigma^{>}$, Σ^{R}, are obtained from the Σ^{p} by Langreth relations [14, 22]. We consider a system like the one shown in Fig. 1. It has three parts: the left (L) and right (R) reservoirs, and a central region - the device (D) that can have, in principles, arbitrary size and shape. Also both reservoirs could have arbitrary shapes but, as it is assumed, the electrons in these sub-systems have well-defined temperatures and chemical potentials. A general assumption is that there is no coupling between reservoirs. All the above assumptions lead to the following Hamiltonian of the system in the matrix form

$$H = \begin{pmatrix} H_{LL} & H_{LD} & 0 \\ H_{DL} & H_{DD} & H_{DR} \\ 0 & H_{RD} & H_{RR} \end{pmatrix}. \tag{4}$$

If the hamiltonian (4) is that of a non-interacting system then basically H is a matrix. The infinite dimensionality of the problem can averted with the use of GF techniques. The retarded single-particle GF is the inverse of H defining the retarded single-particle GF G^R as the inverse of $[(\varepsilon + i\eta) - H]$ where $\eta = 0+$. Similarly, the advanced single-particle GF G^A is the inverse of $[(\varepsilon - i\eta) - H]$. It is then possible to express the GF in the device region D as

$$G^{R,A}_{DD} = \left[(\varepsilon \pm i\eta) - H_{DD} - \Sigma^{R,A}_{B} \right]^{-1}, \tag{5}$$

where $\Sigma^{R,A}_{B} = \Sigma^{R,A}_{L} + \Sigma^{R,A}_{R}$ is self-energy due to the coupling to both contacts. Here $\Sigma^{R,A}_{L} = H_{LD} g^{R,A}_{L} H_{DL}$ is self-energy due to the coupling to the left contact and $g^{R,A}_{L}$ is the surface GF of the left semi-infinite contact. We have a similar expression for the right contact. We note that due to the coupling to the contacts the poles of the GF are not real which is the signature of an open system. The self-energies $\Sigma^{R,A}_{B}$ are thus determined by the boundary conditions imposed to the device. In general, Σ^{R}_{B} gives the spectrum shift and loss in the device due to coupling of the device to the contacts and $\Sigma^{<}_{B}$ accounts for the in-scattering from the contacts to the device. Following Langreth rules the Dyson equations for $G^{<}$ and G^{R} in the device in the matrix form (after Fourier transform with respect to $t - t'$) are

$$G^{<}_{DD} = G^{R}_{DD} \Sigma^{<}_{B} G^{A}_{DD} \tag{6}$$

$$G^{R}_{DD} = \left(\varepsilon + i\eta - H_{DD} - \Sigma^{R}_{B} \right)^{-1}. \tag{7}$$

The goal of the calculation for any electron device is the expression of current with respect to some boundary conditions imposed to contacts, for example a bias voltage. The form of the current injected from the left contact in terms of the GFs is

$$J_L = \frac{2e}{\hbar}\int \frac{d\varepsilon}{2\pi} Tr\left[\Sigma_L^{>}(\varepsilon)G_{DD}^{<}(\varepsilon) - \Sigma_L^{<}(\varepsilon)G_{DD}^{>}(\varepsilon)\right]. \tag{8}$$

Here *Tr* means the trace of an operator. A similar expression is obtained for charge density just by using $G^{<}$ in the device. In the next two sections we will show two examples of device calculations in which the NEGF theory has both semi-empirical and ab-initio implementations.

3. RESONANT TUNNELING THROUGH DOUBLE BARRIER STRUCTURE

Electron transport through resonant tunneling (RT) has been proposed in the context of multi-layers semiconductor heterostructures by the pioneering work by Chang, Esaki and Tsu [23], who called this configuration resonant tunneling diode (RTD). In RTD structures there is a bias dependence of the tunneling current through the double-barrier structure showing a negative differential resistance (NDR) as a result of resonant tunneling due to the alignment between the electron states in the emitter and the quasi-bound state accommodated within the barriers. Experimental results reported in Ref. 24 showed only a weak signature of NDR current-voltage (I-V) characteristics at low temperatures. In the mid 1980's a large NDR was obtained [24] paving the way to the practical nonlinear solid state devices based on purely quantum phenomena.

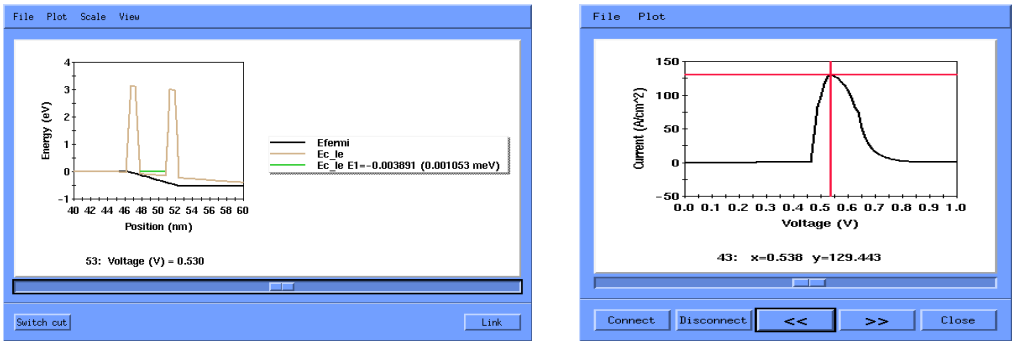

Fig.2. The RTD structure (left) and its I-V characteristics (right). The peak current is obtained for a bias voltage of 0.538 V, which is the resonance condition of the quantum level in the well with the bottom of the conduction band of the emitter

In Fig. 2 we show an RTD structure with barriers of 11.3 Å width and a 31.1 Å wide well with 2X10^{15} cm^{-3} doping. The RTD is clad with 48.9 Å undoped spacer and high doping (1x10^{18} cm^{-3}) contact layers (emitter and collector). The physical parameters of the structure are those found in Ref. 25, namely the effective mass of carriers (electrons) is $m^*=0.19\ m_0$ with m_0 the free electron mass and the barrier height is approximately 3 eV. The calculations were performed with NEMO for a one band model of the RTD with no incoherent scattering. The charging effects are obtained in the local density approximation. The I-V characteristics shows an NDR behavior with the maximum of current density obtained at the resonance between the lowest quasi level in the well and the bottom of the conduction band. (Fig. 2)

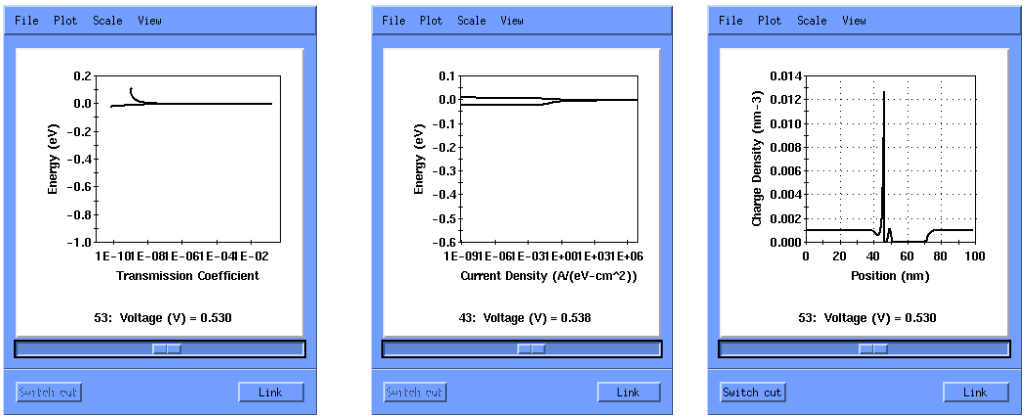

Fig.3. Energy resolved transmission coefficient (left) and current density (center); position dependent charge density is depicted on the right-side hand. All three quantities are obtained under a bias voltage of 0.538 V.

In Fig. 3 we present the energy resolved transmission coefficient and current density as well as the charge density at resonance. The resonant behavior is reflected in both the transmission coefficient and in current density. Also, at resonance there is a charge accumulation in the well. In addition, in the emitter region close to the barrier there is also a charge accumulation due the quasi-bound state formed in the undoped spacer in the emitter. When incoherent scattering phenomena like those due to phonons, alloy disorder, and interface roughness are considered the NDR quality degrades together with the resonant tunneling and the ratio between the peak current and the valley current (i. e., the post-resonance current) decreases drastically [26]. Moreover, the quasi-bound state in the emitter may play a major role in phenomena like hysteresis [27,28]. However, the NEGF theory provides an invaluable insight into the physics of RT even just considering the coherent transport by taking into account the charging and doping effects of electron transport in RTDs.

4. INELASTIC TRANSPORT THROUGH A MOLECULE BRIDGING TWO METALLIC CONTACTS

Transport in single molecules sandwiched between two metal surfaces depends drastically on the interface between the metal surface and the molecule. Therefore, an ab-initio approach to transport seems to be appropriate to describe the contact molecule and metal. Moreover, IETS has become a method of detecting by electrical means what type of molecules is bonded to an electrode [29]. By IETS it is assessed the electron transport in the presence inelastic phonon scattering and the IETS signal is basically the ratio $\left(d^2I/dV^2\right)/\left(dI/dV\right)$. We studied the inelastic transport in benzene-1,4-dithiol (BDT) molecule sandwiched between gold electrodes. The calculations are performed with TranSIESTA [17,18] and its extension to inelastic transport [19]. The results are presented in Fig. 4. The calculations revealed four eigen-channels for electron transport (Fig. 4-left). These eigen-channels provide a complete picture of the electron transport [30]. In addition to that, the IETS signal shows the signature of the vibrational levels of BDT molecule. The arrows shown in Fig. 4-right are consistent with the experimental data from a recent work [31]. Thus the ab-initio calculations may become accurate, but they can be hardly applied to systems encompassing hundreds of atoms. On the other side, regarding the system size, the semi-empirical methods can reach the milestone of thousand of atoms although with lesser accuracy. To finish this section, we may assert that the ab-initio NEGF methods provide not only qualitative insights into the physics of electron transport at molecular level but also quantitative assessments that can be compared with experiments.

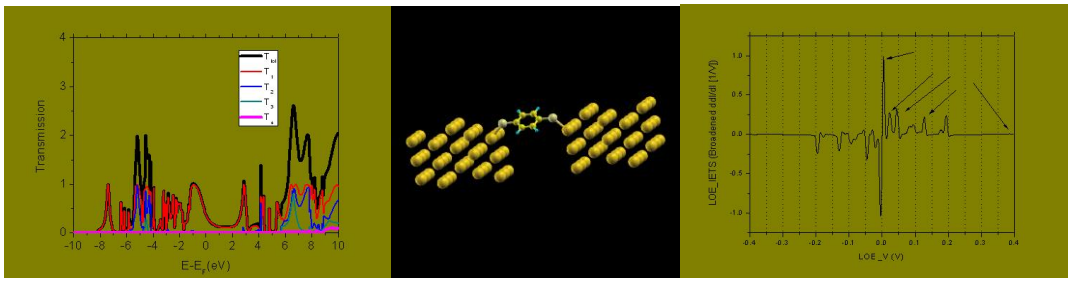

Fig.4. Transmission eigen-channels (left) of a BDT molecule (middle) and the IETS signal (right).

5. CONCLUSIONS

We presented the modern formalism of the non-equilibrium Green's function that is currently applied in numerous studies regarding nanometer size electron devices. After a short introduction into this formalism we used it to study nanometer thick resonant tunneling devices and the inelastic electron transport in a molecule sandwiched between two gold electrodes. Analysis of the results showed that the non-equilibrium Green's function approach may reveal many physical insights which can be used in developing and designing quantum devices.

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