Quantum Transport Study on "Gold-Benzene Dithiol-Gold" Electrical Conductance: DFT-NEGF Calculations

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Abstract

One of the most important problems in molecular electronics is to calculate the quantum transport and the contact resistance of the molecular coupling to the metal electrodes. The well-known approach to attaching nanostructures to external electrodes is to use thiolate bonds. Our computational method is based on solving the Non-equilibrium Green's function (NEGF) in conjunction with density functional theory (DFT). We use the quantum Atomistic Tool Kit (ATK) program package. We calculate quantum transmission spectra, the projected device density of states (PDDOS), the IV characteristics, and the electrical conductance of the two-probe nanostructure: gold- benzene dithiol-gold. The electronic structure and energy bandgap of a two-probe open system is achieved via PDDOS. Besides, the electrical behavior shows that the conductance of this nanostructure is nonlinear.

Keywords: Quantum transport, *Non-equilibrium Green's function (NEGF), density functional theory (DFT), IV characteristics.*

1. Introduction

Quantum transport of electric charge carriers is an inevitable necessity in nanoelectronics [1]. The electrical behavior of an atomic structure is based on the exact calculation of the amount of electric current for different applied voltages. Finding electric current in structures with nanometer dimensions requires calculations of carrier quantum transport [2]. In this type of transport, we see the carriers ballistic transport and takes place through the path from the right electrode to the left electrode without colliding with other particles. Quantum transport in nanostructures at low temperatures differs from what Ohm's laws predict. Quantization of electrical conductance in coherent and ballistic transport is one of the quantum transport behaviors. Presently, the most common method for calculating coherent transport is to solve non-equilibrium Green's equations (NEGF) with the Landauer-Buttiker formalism [3]. In this formulism, the coherent electrical conductivity of a sample whose Hamiltonian is already described in Density Functional theory (DFT) can be calculated. In a more advanced formalism called the Keldysh method, the effect of adding interactions can also be involved in solving non-equilibrium Green equations and solving NEGF equations in an non-coherent way [4]. A practical example is the non-coherent solution and the effect of the addition of interactions, light interaction or the effect of incident photon energy on quantum transport. One of the advantages of Keldysh method is the possibility of calculating the behavior of the time variable system. Spin properties can also be studied in this method.

Preceding reports on calculating the quantum transport of nanostructures can be found in reliable journals [5-7]. Solutions of non-equilibrium Green equations for quantum transport calculations with tight

binding Hamiltonian have also been performed. Though tight binding-based computations require fewer computer resources and respond faster, the need to determine semi-Empirical parameters for the nanostructured tight-binding model means that tight binding-based computations are not as accurate as first principles calculations [8-10].

In the current paper, the results of quantum computations with Hamiltonian based on density functional theory (DFT) for solving non-equilibrium Green equations are reported, resulting from the circadian work of high performance computers. The structure under study is a carbon structure called benzene dithiol (BDT), which is attached to gold electrodes to apply an external potential to establish an electric current and achieve its electrical behavior. In addition to extracting the IV characteristic of nanoelectronic devices, the results of this study can be used to design and analyze biosensors involved with vital molecules or to study electrically conductive polymers.

2. Theory and Model

In this study, quantum transport calculations are performed in ATK software with non-equilibrium Green functions (NEGF) according to Keldysh formalism in combination with self-consistent field theory (SCF) [11]. The self-consistent field theory used in this software based on Hamiltonian is similar to DFT. It consists of such elements as the kinetic energy of the electrons, the Hartree Potential, the exchange-correlation Potential (xc), the potential due to the charge of the nucleus of the atoms, and the externally applied potential component. These potentials look similar to what we had in DFT, but the density of electrons used to calculate these potentials is derived from non-equilibrium Green functions, and the equation solution under equilibrium conditions is no longer used to find the density of electrons. Likewise, due to the unusability of the variational principles in non-equilibrium conditions, the NEGF-SCF equations are no longer solved based on the method of minimum total energy, similar to what we had in DFT. Somewhat, the density of electrons is calculated based on the direct solution of Hamilton, albeit in a self-consistent cycle. Therefore, the NEGF-SCF non-equilibrium Green equations method is substantially and qualitatively different from the ground state DFT method. In other words, the NEGF-SCF method is not a ground-state method because, in this method, the electron density tensor is calculated as non-equilibrium and non-variance. Despite this fundamental difference, the litratures also refer to NEGF-SCF as NEGF-DFT, simply because the SCF cycle is similar to DFT [12-14]. However, it should not be unnoticed that the DFT used in the NEGF-DFT title differs in content from that in the DFT equilibrium, although both have a similar Hamiltonian. Figure 1 shows the two-probe atomic structure designed for quantum transport calculations. This figure shows the “gold-benzene dithiol-gold” atomic structure designed by ATK software. Gold atoms are shown with large golden spheres, sulfur atoms yellow spheres, carbon atoms smaller brown spheres, and hydrogen atoms very small pink spheres that surround the benzene molecule. The role of hydrogen atoms has been used to passivate carbon atoms.

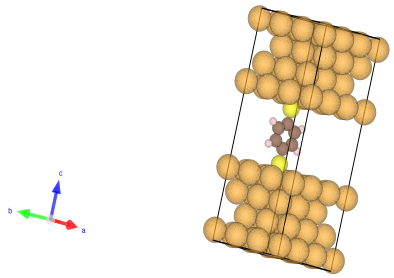

Fig. 1. Two-probe atomic structure that used for quantum transport study

3. Results and Discussion

Based on the structure designed in Figure 1 and by LDA functional [15], we performed SCF calculations and succeeded in extracting the energy spectrum of the quantum transmission coefficients of the two-probe structure discussed in Figure 2. A sharp peak is observed in this spectrum for the quantum transmission coefficient for energy +2.8 eV. The value of *T(E)* in this peak is grater than one (>1.5), but we know the probability of carrier transport in any channel is less than one. The physical interpretation of this result relates to the cumulative nature of the total transmission coefficient, *T(E)*. Because the number of quantum transmission channels that can be used between the two electrodes is more than one channel, the total transmission coefficient can be larger than one.

We also see another wide peak at -1.1eV energy in Figure 2. Based on these peaks, it can be predicted that if the external bias applied to this device is less than 1.0V and the bias window does not include these peaks, no significant electric current will be generated.

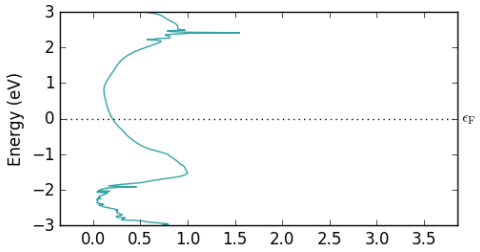

Fig. 2. Quantum Transmission spectra, T (E), of studied two probe nanostructure: Au-DTB-Au

By opening the bias window to ± 2V, the first current increase will occur, and if we increase the bias window to ± 3V, we will see the second step of increasing the device current. The completely nonlinear behavior of the electrical conductance of the studied structure is obvious.

To better analyze the spectrum of the quantum transmission coefficient shown in Figure 2, we have calculated the density of PDDOS states. From the density of states, the electronic structure of the studied device is obtained. In open two-probe systems, we use the DOS spectrum to study the electronic properties instead of using the energy band structure. The energy bands structure is used to study periodic systems and isolated systems. Figure 3 shows the PDDOS of the studied device states in the ±3eV energy window.

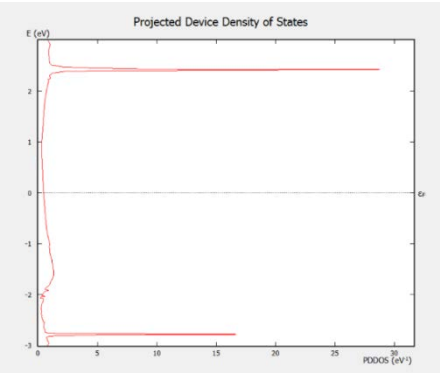

Fig. 3. The projected device density of states (PDDOS) of studied nanostructure: Au-DTB-Au

Then, we perform a IV analysis on the designed structure, and for the bias voltage range ± 5.6V with a step of 0.4V, we have drawn the electrical characteristics of the structure in Figure 4. The values of electric current in this figure are listed in microamperes.

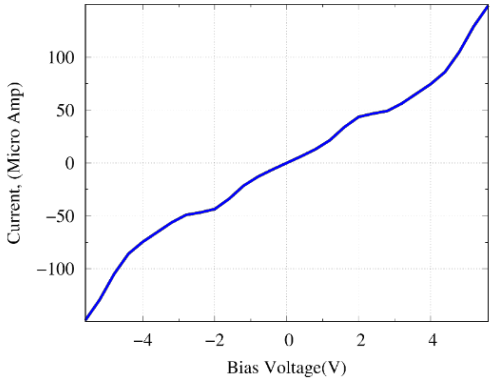

Fig. 4. The IV-characteristics of studied two probe nanostructure: Au-DTB-Au

Another curve considered in this work is the electrical conductance of the structure, which is calculated from the current-to-voltage ratio and is illustrated in Figure 5. We have obtained the electrical conductance or G (E) in nano-Siemens and the bias range of ± 2.0V. In nanostructures, by changing the amount of bias applied, the number of channels in the bias window changes, and we see a change in the electrical conductance of the device. Regarding the symmetry of the structure in terms of left and right electrodes, the shape of the electrical conductance curve obtained for the structure is symmetrically obtained in terms of the polarity of the applied voltage and the electrical conductance behavior in negative and positive biases is not different. We have obtained the maximum amount of electrical conductance of less than 35µS.

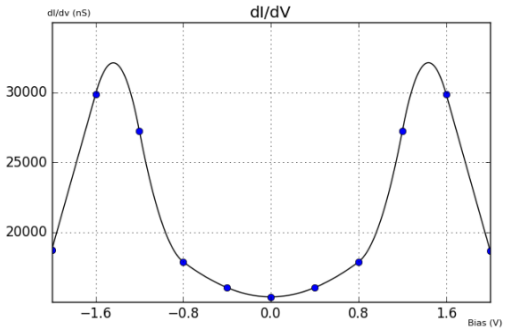

Fig. 5. The electrical conductance of studied two probe nanostructure: Au-DTB-Au

Conclusions

In molecular electronics or nanoelectronics, work begins with designing atomic bonds between metal contacts and the nanostructure under study. Based on the number of quantum transmission channels that can be used between the two electrodes, the total transmission coefficient can be cumulatively larger than one. We know the probability of carrier transport in any channel is less than one. By calculating the quantum transmission coefficient spectrum, the way is opened to calculate the density of states, PDDOS, and detailed information is obtained from the electronic structure of the two-probe nano-device. In this regard, we can then calculate the electric current passing due to the application of external potential and draw the IV curve of the nano-device. From the IV curve and by the derivative of $\frac{di}{dv}$, the electrical conductance of the nanostructure is obtained, and in practice, we get a complete understanding of the electrical behavior of such a structure. In this paper, we have done such a computational process as above, based on solving the non-equilibrium Green equation using Hamilton density theory, and obtained the electrical behavior of the “gold-benzene dithiol-gold” nanostructure

with ATK software. Surely, this computational work can be a context for developing future work for various applications such as biosensors, solar cells, nano-optoelectronics, etc.

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References

[1] S. Datta, Lessons from Nanoelectronics vol. Volume 5, World Scientific, 2017.

[2] J. Taylor, H. Guo, and J. Wang, "Ab initio modeling of quantum transport properties of molecular electronic devices", Physical Review B, vol. 63, no. 24, pp. 24540701-24540713, 2001.

[3] M. Pourfath, The Non-Equilibrium Green's Function Method for Nanoscale Device Simulation, 1st ed., Springer-Verlag Wien, 2014.

[4] H. Guo, L. Liu, Y. E. Zhu, S. Bohloul, V. Michaud-Rioux, Y. Hu, J. Taylor, and L. Wan, "Nanodcal: an LCAO implementation of density functional theory (DFT) within the Keldysh nonequilibrium Green's function formalism (NEGF)," ed. CANADA: NanoAcademic Technologies Inc., 2001.

[5] A. Hernández, V. M. Apel, F. A. Pinheiro, and C. H. Lewenkopf, "Quantum electronic transport: Linear and nonlinear conductance from the Keldysh approach", Physica A: Statistical Mechanics and its Applications, vol. 385, no. 1, pp. 148-160, 2007.

[6] R. Gutiérrez, S. Mandal, and G. Cuniberti, "Quantum Transport through a DNA Wire in a Dissipative Environment", Nano Letters, vol. 5, no. 6, pp. 1093-1097, 2005.

[7] V. N. Likhachev and G. A. Vinogradov, "The Green's Function in the Problem of Charge Dynamics on A One-Dimensional Lattice with An Impurity Center", Theoretical and Mathematical Physics, vol. 196, no. 1, pp. 1018-1027, 2018.

[8] F. Ostovari and M. K. Moravvej-Farshi, "Photodetectors with zigzag and armchair graphene nanoribbon channels and asymmetric source and drain contacts: Detectors for visible and solar blind applications", Journal of Applied Physics, vol. 120, no. 14, pp. 144505-144511, 2016.

[9] W. J. Jeong, J. Seo, and M. Shin, "Efficient TB-NEGF simulations of ultra-thin body tunnel FETs," in 2016 International Conference on Simulation of Semiconductor Processes and Devices (SISPAD), 2016, pp. 81-84.

[10] A. Dinarvand, V. Ahmadi, and G. Darvish, "Asymmetric bilayer graphene nanoribbon MOSFETs for analog and digital electronics", Superlattices and Microstructures, vol. 93, pp. 202-213, 2016.

[11] S. Smidstrup, T. Markussen, P. Vancraeyveld, J. Wellendorff, J. Schneider, T. Gunst, B. Verstichel, D. Stradi, P. A. Khomyakov, et al., "QuantumATK: an integrated platform of electronic and atomic-scale modelling tools", Journal of Physics: Condensed Matter, vol. 32, no. 1, p. 015901, 2019.

[12] M. Balarastaghi, V. Ahmadi, and G. Darvish, "Electro-optical properties of new structure photodetectors based on graphene nanoribbons: an ab initio study", Journal of the Optical Society of America B, vol. 33, no. 11, p. 2368, 2016.

[13] N. Papior, N. Lorente, T. Frederiksen, A. García, and M. Brandbyge, "Improvements on non-equilibrium and transport Green function techniques: The next-generation transiesta", Computer Physics Communications, vol. 212, pp. 8-24, 2017.

[14] M. Balarastaghi, V. Ahmadi, and G. Darvish, "Electro-optical properties of new structure photodetectors based on graphene nanoribbons: an ab initio study", Journal of the Optical Society of America B, vol. 33, no. 11, pp. 2368-2373, 2016.

[15] J. P. Perdew and A. Zunger, "Self-interaction correction to density-functional approximations for many-electron systems", Physical Review B, vol. 23, no. 10, pp. 5048-5079, 1981.