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Electronic and thermal properties of Biphenyl molecules

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ABSTRACT

Transport properties of a single Biphenyl molecule coupled to two contacts are studied. We characterise this system by a tight-binding Hamiltonian. Based on the non-equilibrium Green's functions technique with a Landauer–Büttiker formalism the transmission probability, current and thermoelectrical power are obtained. We show that the Biphenyl molecule may have semiconductor behavior for certain values of the electrode–molecule–electrode junctions and different values of the angle between the two rings of the molecule. In addition, the density of states (DOS) is calculated to compare the bandwidths with the profile of the transmission probability. DOS allows us to explain the asymmetric shape with respect to the molecule's Fermi energy.

1. Introduction

The molecular electronics has had a remarkable progress during the past decades, in particular by the synthesis of molecules and assembling them to electronic devices, getting new applications in nanoscale systems. The transport properties have been determined, in order to use molecular systems in practical devices such as in organic solar cells, organic light-emitting diodes, molecular circuits, and organic field-effect transistors, just to mention a few [1–4].

These systems may be based on single-molecules and have been studied from both, the theoretical and experimental point of view. In fact, it has been found that the mechanisms of different electronic behaviors depends on the molecule's geometry or the external agents [5–8]. The deep understanding of the transport and thermal properties of aromatic molecules helps us in the development of molecular devices, and for that reason we focus in the present work on the study of the electronic as well as thermodynamic properties of the Biphenyl molecule ($C_{12}H_8$) coupled to two semi-infinite leads.

The aim of this work is to explore both, the electrical and the thermal fluctuations, in the transport through the Biphenyl molecule. In particular, we study these properties by changing the geometry of the molecule and by assuming different regimes of the coupling between the molecule and the contacts. The molecule is modeled by a tight-binding Hamiltonian with a nearest-neighbor approximation. The calculations of the transport properties based on electron transfer and heat transport are performed using non-equilibrium Green's functions techniques inside of a discrete space. In particular, we use a decimation procedure or a real-space renormalisation scheme [9–11]. The transmission probability, thermoelectrical power and current as a function of the bias voltages are obtained following the Landauer–Büttiker formalism [12]. In

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addition, we calculate the density of states (DOS) to compare the bandwidths with the profile of the transmission probability and the Seebeck coefficient in the limit of strong and weak couplings. We show a good agreement between both methods.

The manuscript is organised as follows: In Section 2, the theoretical model is introduced and the system's transport quantifiers are described. In Section 3 numerical simulations are performed and the results are explained. Finally, a summary is given in Section 4.

2. Theoretical model

To describe the Biphenyl aromatic molecule as an electronic device, the molecule is connected to contacts at both ends. Each ring consists of $n = 6$ carbon atoms. The set-up is depicted in Fig. 1.

The Hamiltonian of the total system in the a tight-binding approximation is given by:

$$\widehat{H} = \widehat{H}_{mol} + \widehat{H}_e + \widehat{H}_c, \tag{1}$$

where $\widehat{H}_{mol}$ is the Hamiltonian of the Biphenyl molecule without contacts which can be written as:

$$\widehat{H}_{mol} = \sum_i \epsilon \hat{c}_i^\dagger \hat{c}_i + \sum_i v\left[\hat{c}_{i+1}^\dagger \hat{c}_i + \hat{c}_i^\dagger \hat{c}_{i+1}\right] + \sum_j \epsilon \hat{c}_j^\dagger \hat{c}_j + \sum_j v\left[\hat{c}_{j+1}^\dagger \hat{c}_j + \hat{c}_j^\dagger \hat{c}_{j+1}\right] + t\left[\hat{c}_4^\dagger \hat{c}_7 + \hat{c}_7^\dagger \hat{c}_4\right]. \tag{2}$$

Here, ϵ is the electron energy per site or per carbon atom, $c_i^\dagger$ is the creation operator of an electron at site i in the ring 1, $c_j^\dagger$ is the creation operator of an electron at site j in the ring 2, v is the hopping parameter between carbon atoms and t is the torsional potential. The Hamiltonian which represents the electrodes, $\widehat{H}_e$, is given by:

$$\widehat{H}_e = \sum_{k_R} \epsilon_{k_R} \hat{d}_{k_R}^\dagger \hat{d}_{k_R} + \sum_{k_L} \epsilon_{k_L} \hat{d}_{k_L}^\dagger \hat{d}_{k_L}. \tag{3}$$

Finally, the Hamiltonian that describes the interaction between the contacts and molecule can be cast in the form:

$$\widehat{H}_c = \sum_{k_R} \Gamma_R \hat{d}_{k_R}^\dagger \hat{c}_1 + \sum_{n_L} \Gamma_L \hat{d}_{k_L}^\dagger \hat{c}_N + h.c., \tag{4}$$

where the operator $\hat{d}_{k_{L(R)}}^\dagger$ is the creation operator of an electron in a state $k_{L(R)}$ with energy $\epsilon_{k_{L(R)}}$, while $\Gamma_{R(L)}$ is the coupling energy between the right (left) lead with the Biphenyl molecule.

The thermodynamic and electronic properties of a single Biphenyl molecule coupled to two contacts are calculated through the Landauer–Büttiker formalism [12] based on the Green's function techniques. In particular, the Green's functions are computed within a real-space renormalisation approach. This method allows us to obtain all the electronic information of the molecular system [13]. In this scenario, the resultant molecular system is reduced to an unidimensional system with effective energy sites and effective couplings between them as is shown in Fig. 2.

The Green's function of the aromatic molecule coupled to the leads is calculated using the Dyson equation, which is given by

$$G = G_0 + G(\Sigma_L + \Sigma_R)G_0, \tag{5}$$

where G^0 is the bare Green's function of the isolated aromatic molecule and Σ_L and Σ_R are the self-energies of the left and right lead, respectively. The detail of the renormalization method and analytical expressions of the Green's functions are given in the Appendix. The transmission probability in the reduced system can be calculated within the Fischer–Lee relationship [14]:

$$T(E) = \Gamma_{11}^L \Gamma_{NN}^R \left|G_{1N}^r\right|^2, \tag{6}$$

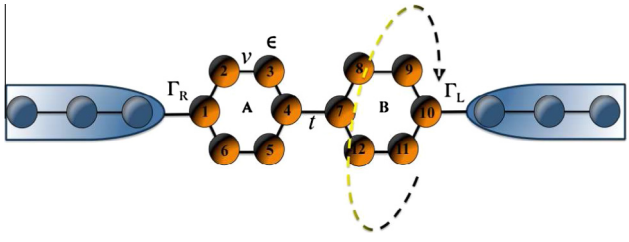

Fig. 1. Biphenyl aromatic molecule.