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Shot noise and thermopower in aromatic molecules



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HIGHLIGHTS

- The quantum transport of aromatic molecules connected to two semi-infinite leads is studied.
- We use Green's function techniques within a real space renormalization scheme.
- We calculate the transmission probability, the shot noise and the thermopower.
- Our results show different transport regimes as a function of the coupling and the length of chains.

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ABSTRACT

The quantum transport properties of finite homogeneous chains of aromatic molecules composed of one, two or several benzene rings connected to two semi-infinite leads are studied. We study these molecules based on the tight-binding approach. The calculation of the transport properties is performed using Green's function techniques within a real space renormalization scheme. In particular, we focus on the transmission probability, on the noise power of current fluctuations and on the thermopower. Our results show different transport regimes for these molecular systems as a function of the coupling intensities and the length of chains.

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1. Introduction

In recent decades, the nanoscience has developed potential applications in technology and built electronic devices with different molecular configurations. In particular, the nanostructures based on single-molecules are devices that have been studied from both theoretical and experimental point of view, since they behave as insulators, semiconductors or conductors depending on the geometry or the external effects [1–8].

In this framework, the study of transport properties in molecular systems composed of individual molecules, such as benzene or configurations of benzene rings, has played an important role in building new nano-electronic devices designed to be used as molecular electronic transistors. This technology is based on electron transfer and presents interesting properties on heat transport or magnetoresistance, just to mention a few [9–12].

The mechanism for the study of the electron transport through aromatic molecules is based on placing the molecules between metal contacts. The current through the molecular system is in general a non-linear function of the bias voltages or applied gate voltage. Due to this nonlinear behavior, it is possible to observe quantum fluctuations in the transport properties of aromatic molecules [13,14]. In fact, in the absence of scattering processes, these fluctuations are known as the noise power spectrum, which in the steady state is described by the shot noise. The noise power provides an important information about the electronic correlation by means of the Fano factor (F), which indicates whether the magnitude of the noise reaches a Poisson ($F = 1$) or sub-poisson ($F < 1$) limits [15,16].

On the other hand, a magnitude that can provide additional information about the electronic transport is called the Seebeck effect [17–20], which relates the amount of potential difference developed in a material with the temperature. This effect in molecular systems is produced when a voltage difference between the contacts due to temperature difference exists between the source and the drain. In addition, a large set of benzene rings can be formed as molecular wire. These wires are specially important, because they can exhibit

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thermopower due to resonant transmission through molecular orbitals.

Moreover, if the transmission is not a constant, then it produces a finite current for a temperature difference in the two electrodes, even in the absence of any applied bias. Depending upon the shape of this transmission function we can calculate the Seebeck coefficient (S_c), and consequently determine the fermi level E_f with respect to the position of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO). The positions of the HOMO and LUMO levels are given by the resonances in the transmission spectrum at the edges of the band gap. Hence, one can determine the Fermi level, since it is inside the HOMO–LUMO gap [12]. To clarify this sentence, let us remark that the E_f is given when the number of states is equal to the number of electrons in the molecule. Hence, if the molecule is connected to the contacts, the Fermi level does not remain neutral. Consequently, it can pick up a fractional of charge depending on the work function of contact, and the transferred charge between molecules to the contact is usually much less than the unity. If the fraction is equal to $+1$, the Fermi energy must be on the LUMO, while if the fraction is -1 , it must be on the HOMO. Otherwise, for values in between ± 1 , the E_f should be somewhere inside the HOMO–LUMO gap.

The aim of this work is to explore both the electrical and the thermal fluctuations in the transport through segments of aromatic molecules. In particular, we study from one to seven benzene aromatic rings placed between two metal contacts. These molecules are modeled using a tight-binding Hamiltonian within a nearest-neighbor approximation, and the transport properties are calculated through Green's functions formalism. We focus on the shot noise and on the thermopower in such molecules. We find that benzene molecules display characteristics of conductor, semiconductor and insulating devices depending on the parameters. The manuscript is organized as follows: In Section 2, we introduce the model based on a tight binding Hamiltonian approximation. In Section 3, we describe the theoretical method and calculate Green's functions. In Section 4, we present the results and perform the corresponding discussions. Finally, we summarize the work in Section 5.

2. Models

In order to describe the aromatic molecule as a electronic device we have adopted homogeneous chains with N benzene's rings. In particular, we have connected one, two or more benzene's rings to the contacts as we can observe in Fig. 1(a). Each ring is characterized by having $n=6$ carbon atoms.

The full system is described by a tight-binding Hamiltonian, given by

$$H = H_{AR} + H_L + H_I, \quad (1)$$

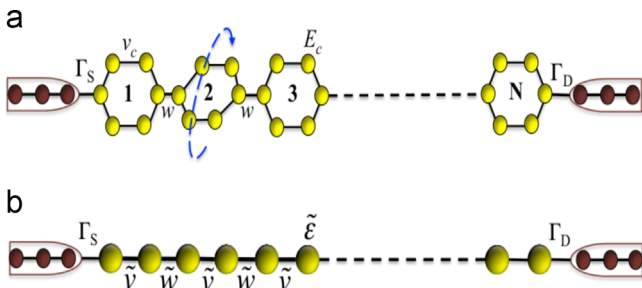


Fig. 1. (a) Aromatic molecules and (b) effective molecular chains. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this article.)

where H_{AR} corresponds to the Hamiltonian of the aromatic molecule embedded between two electrodes, and is given by

$$H_{AR} = \sum_i v_c (c_i^\dagger c_{(i+1)} + c_{(i+1)}^\dagger c_i) + w \sum_i (c_i^\dagger c_{(i+1)} + c_{(i+1)}^\dagger c_i) + \sum_i E_c c_i^\dagger c_i \quad (2)$$

where c_i^\dagger is the creation operator of an electron at site i , v_c is hopping between carbon atoms, w is the hopping between the benzene's rings along the molecule and E_c is the energy of carbon atom.

On the other hand, H_L represents the leads and H_I their interaction with the molecule, given by

$$H_L = \sum_{k_L} \epsilon_{k_L} d_{k_L}^\dagger d_{k_L} + \sum_{k_R} \epsilon_{k_R} d_{k_R}^\dagger d_{k_R} \quad (3)$$

and

$$H_I = \sum_{k_L} \Gamma_S d_{k_L}^\dagger c_1 + \sum_{k_L} \Gamma_D d_{k_R}^\dagger c_N + h.c., \quad (4)$$

where the operator $d_{k_{L(R)}}^\dagger$ is the creation operator of an electron in a state $k_{L(R)}$, with energy $\epsilon_{k_{L(R)}}$, while $\Gamma_{S(D)}$ is the coupling between each lead (Source–Drain) with the aromatic molecule.

3. Method

We study the transport properties through aromatic molecules by using the Landauer–Büttiker formalism [13–15], based on Green's function techniques within a real-space renormalization approach (decimation procedure) [21,22]. For complex systems the decimation method allows us to solve a set of infinite eigenvalue equations, which is used to compute Green's functions. This method is widely applied in pure and disordered systems [23,24], in which the renormalized real space contains all the electronic information of the molecular system. In addition, in the case of tight-binding models, the decimation process produces that the information about the couplings is substituted by effective couplings in other sites, such that the boundaries are remained fixed.

Green's function of the aromatic molecules coupled to the leads is calculated by using the Dyson equation given by $G = G^0 + G^0 (\Sigma_L + \Sigma_R) G$, 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 leads, respectively. The transmission probability can be obtained by using the Fischer–Lee [13] relationship, which is given by

$$T(E) = \text{Tr}[\Gamma^L G^r \Gamma^R G^a], \quad (5)$$

where $\Gamma^{L(R)} = i(\Sigma^{L(R)} - \Sigma^{L(R)\dagger})$ is the spectral matrix density of the left(right) lead; and (G^r, G^a) are the retarded and the advanced Green's function, respectively.

As we can see, we transform the aromatic molecule representation into an effective one-dimensional chain of sites [see Fig. 1(b)], obtaining renormalized Green's functions with effective inter-site couplings, which contain all the information of the planar aromatic molecule. Then, the transmission probability can be written as

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

where Γ_{11}^L and Γ_{NN}^R depict the element $\{1,1\}$ and $\{N,N\}$ of the effective spectral matrix density of the left and right leads, respectively. Also, Green's function G_{1N}^0 , G_{NN}^0 and G_{11}^0 can be analytically determined by using the renormalization techniques, where N represents the effective atomic site. In what follows, we take $\Sigma^L = \Sigma^R = -i\Gamma/2$ and $G_{NN}^0 = G_{11}^0$.

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