Quantum transport through a molecule coupled with a mesoscopic ring

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Received 31 May 2006
Accepted for publication 9 September 2006
Published 5 December 2006
Online at stacks.iop.org/PhysScr/75/62

Abstract
Transport through a molecule sandwiched between two metallic electrodes and coupled with a mesoscopic ring that threads a magnetic flux $\phi$ is studied. An analytic approach for electron transport through the molecular bridge system is presented. The electronic transport properties are discussed in two aspects: (i) the presence of an external magnetic field and (ii) the strength of the molecule to electrodes coupling.

PACS numbers: 73.23.-b, 73.63.-b, 81.07.Nb

1. Introduction

Much progress in nanofabrication of quantum devices has allowed one to study the electron transport through molecules in a very controllable way and molecular electronics have attracted much more attention since molecules constitute promising building blocks for a future generation of electronic devices. The electron transport through molecules was first studied theoretically in 1970 [1]. Since then several experiments [2–5] have been performed through molecules placed between two metallic electrodes with few nanometres separation. The operation of such two-terminal devices is due to an applied bias. Current passing across the junction is a strongly nonlinear function of applied bias voltage and its detailed description is a very complex problem. Complete knowledge of the conduction mechanism at this scale is not well understood yet, but the transport properties of these systems are associated with some quantum effects, such as quantization of energy levels and quantum interference of electron waves. A quantitative understanding of the physical mechanisms underlying the operation of nano-scale devices remains a major challenge in nanoelectronics research.

The aim of this paper is to reproduce an analytic approach based on the tight-binding model to investigate the electronic transport properties through a molecule coupled with a mesoscopic ring. There exist some ab initio methods for the calculation of conductance ($g$) and current ($I$) through the molecular bridge system, as depicted schematically in figure 1. The full system (molecule with ring) is described by a single-band tight-binding Hamiltonian with a non-interacting picture, which is written as

$$H = H_M + H_R + H_{MR}$$

where $H_M = \epsilon_{\text{mol}}$ is the site energy of the molecule and $\epsilon_i$'s are the site energies of the ring. $H_R = \sum_i \epsilon_i c_i^\dagger c_i + \sum_{\langle ij \rangle} t(c_i^\dagger c_j e^{i\phi} + \text{h.c.})$ and $H_{MR} = t_0(c_1^\dagger d_m + \text{h.c.})$ are the terms for the molecule and ring, respectively. The term $H_{MR}$ is the hopping integral between the molecule and the leads.

This paper is organized as follows. In section 2, I introduce the molecular system under consideration and give a very brief description for the calculation of conductance ($g$) and current ($I$) through the molecular bridge system. Section 3 presents the results of conductance–energy and current-voltage characteristics for the bridge system taken into account and lastly, I summarize my results in section 4.

2. The molecular model and a brief description of the theoretical formulation

The system under consideration is a small molecule (with one atomic site) coupled with a quantum ring with $N$ atomic sites and sandwiched with two metallic electrodes, as depicted schematically in figure 1. The full system (molecule with ring) is described by a single-band tight-binding Hamiltonian with a non-interacting picture, which is written as

$$H = H_M + H_R + H_{MR}$$

where $H_M = \epsilon_{\text{mol}}$ is the site energy of the molecule and $\epsilon_i$'s are the site energies of the ring. $H_R = \sum_i \epsilon_i c_i^\dagger c_i + \sum_{\langle ij \rangle} t(c_i^\dagger c_j e^{i\phi} + \text{h.c.})$ and $H_{MR} = t_0(c_1^\dagger d_m + \text{h.c.})$ are the terms for the molecule and ring, respectively. The term $H_{MR}$ is the hopping integral between the molecule and the leads.
Figure 1. A schematic view of a molecule coupled with a mesoscopic ring pierced by a magnetic flux $\phi$ and the molecule is sandwiched between two metallic electrodes.

The characteristic behaviour of conductance as a function of energy in the weak-coupling limit is shown in figure 2, where the solid and dotted curves respectively correspond to the ring ($N = 15$) with $\phi = 0$ and 0.3. Conductance vanishes for almost all energy values, except at resonances where $g = 2T$ (from Landauer formula with $e = \hbar = 1$). The resonance peaks in the conductance spectrum coincide with the eigenenergies of the molecular system and thus the spectrum manifests itself the energy levels of the molecular bridge system. In the presence of magnetic flux, more resonance peaks appear in the conductance spectrum (dotted curve) which reveals that more energy levels appear in the system. The physical reason is that for nonzero values of $\phi$ all the degeneracies of the molecular energy levels are lifted and eventually the system gets more resonating states. Thus by introducing $\phi$, transmission through the bridge system can be controlled.

In the strong-coupling limit, these resonance peaks get substantial widths as observed in figure 3, where the solid and

\[ g = \frac{2e^2}{\hbar} T \]

and the transmission probability of an electron from the source to the drain through the molecular bridge is expressed as [15]

\[ T(E, V) = \text{Trace} \left[ \left( \Sigma_S^0 - \Sigma_D^0 \right) G^0 \left( \Sigma_D^0 - \Sigma_D^0 \right) \right] \]

where $\Sigma_S$ and $\Sigma_D$ are the self-energies due to the coupling of the molecule with the two electrodes. The Green’s function $G$ of the full system is given by the relation,

\[ G = [E - H - \Sigma_S - \Sigma_D]^{-1} \]

where $E$ is the energy of injecting electrons from the source and $H$ is the Hamiltonian of the full system described above. Now all the information regarding the molecule to electrodes coupling is included into the two self energies and is analysed through the use of Newns–Anderson chemisorption theory [12]. The self energies contain real and imaginary parts, where the real parts correspond to energy shift of the energy eigenstates of the molecular system and the imaginary parts give the broadening of these energy levels.

The current passing through the bridge is depicted as a single-electron scattering process between the two reservoirs of charge carriers. Now the current–voltage relation is evaluated from the following expression [15]

\[ I(V) = \frac{e}{\pi \hbar} \int_{E_F - eV/2}^{E_F + eV/2} T(E, V) \, dE \]
Conductance $g$ as a function of energy $E$ in the strong-coupling limit, where the solid and dotted curves respectively correspond to the ring ($N = 15$) with $\phi = 0$ and 0.3.

Current $I$ as a function of bias voltage $V$ in the weak molecule-to-electrodes coupling limit, where the solid and dotted lines respectively correspond to the ring ($N = 15$) with $\phi = 0$ and 0.3.

dotted curves correspond to the same meaning as before. The increment of the resonance width is due to the broadening of the molecular energy levels (contributions come from the imaginary parts of the two self-energies) for strong molecular coupling with the two electrodes.

The scenario of electron transfer through the molecular bridge system is much more clearly observed by studying the current–voltage characteristics. Current is computed by integration of the transmission function $T$ which shows the same variation, differing only by a factor 2 in magnitude, like in the conductance spectrum (figures 2 and 3). The current–voltage characteristic in the weak-coupling limit for the molecular system taken earlier is given in figure 4, where the solid curve corresponds to the current in the absence of $\phi$ and the dotted curve denotes the same for $\phi = 0.3$. The current shows staircase-like behaviour with sharp steps, which is associated with the discrete nature of the molecular resonances (figure 2). It is clearly observed from this figure that in the presence of magnetic flux, the current shows more steps (dotted curves) compared to the zero-flux system, which reveals that more resonance peaks appear in the conductance spectrum for nonzero values of $\phi$. The shape and width of the current steps depend on the width of the molecular resonances since the height of a step in the $I$–$V$ curve is directly proportional to the area of the corresponding peak in the conductance spectrum. Current varies continuously with the applied bias voltage and achieves much higher values in the strong-coupling limit, as shown in figure 5, where the solid and dotted curves correspond to the same meaning as before.

4. Conclusion

In conclusion, I have introduced a parametric approach based on the tight-binding model to investigate the electronic transport properties at absolute zero temperature of a single molecule coupled with a quantum ring that threads a magnetic flux $\phi$. This method is much more flexible compared to other existing theories and can be used to study such transport properties through any complicated molecular bridge system. Electronic conduction through molecular systems is strongly influenced by the flux $\phi$ in the ring and the molecule-to-electrodes coupling strength. These results predict that designing a whole system that includes not only molecules but also molecule-to-electrodes coupling is highly important in fabricating molecular electronic devices.

Acknowledgments

It is a pleasure to thank Atikur Rahman (my best friend) for many helpful comments and suggestions. I also thank Professor S Sil and Professor S N Karmakar for some comments.

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