Quantum Transport in a Biphenyl Molecule: Effects of Magnetic Flux

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Abstract

Electron transport properties of a biphenyl molecule are studied based on the Green’s function formalism. The molecule is sandwiched between two metallic electrodes, where each benzene ring is threaded by a magnetic flux $\phi$. The results are focused on the effects of the molecule to electrode coupling strength and the magnetic flux $\phi$. Our numerical study shows that, for a fixed molecular coupling, the current amplitude across the bridge can be regulated significantly just by tuning the flux $\phi$. This aspect may be utilized in designing nano-scale electronic circuits.

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1 Introduction

In the last few decades, electron transport properties through nano-scale systems\(^1\)\(^–\)\(^4\) have been studied enormously, and recently people are really getting very excited in fabrication of electronic circuits by using single molecule or cluster of molecules\(^5\)\(^–\)\(^6\) due to their unique properties. In the very early days, Aviram and Ratner\(^7\) first predicted theoretically the electron transport through a molecular system, and later many experiments\(^8\)\(^–\)\(^12\) were performed to understand the basic mechanism underlying such transport. In a molecular bridge, the electron transmission is controlled by several important factors and all these effects have to be taken into account properly. The most important issue is probably the quantum interference effects\(^13\)\(^–\)\(^16\) among the electron waves traversing through different arms of the molecule. Another important issue is the molecular coupling to the side attached electrodes.\(^17\) Tuning this coupling, one can control the current amplitude very nicely across the bridge. Similar to these, there are several other factors like the geometry of the molecule,\(^18\) electron-electron correlations,\(^19\) dynamical fluctuations,\(^20\)\(^–\)\(^21\) etc., which provide rich effects in the electron transport.

In the present article we focus on the electron transport properties of a biphenyl molecule, where each benzene ring is threaded by a magnetic flux $\phi$, the so-called Aharonov-Bohm (AB) flux. Quite interestingly we see that, keeping all the other parameters as invariant, the current amplitude across the molecule can be regulated very nicely simply by tuning the magnetic flux $\phi$. Thus we can design an electronic circuit by using the biphenyl molecule and the electron transmission through the circuit can be regulated efficiently just by controlling the parameter $\phi$. This phenomenon can be utilized in designing the future nano-electronic circuits. Here we provide a very simple analytical formulation of the transport problem through the biphenyl molecule using the tight-binding Hamiltonian, and the coupling of the molecule to the side attached electrodes is treated through the Newns-Anderson chemisorption theory.\(^22\)\(^–\)\(^24\) There exist several \textit{ab initio} methods\(^25\)\(^–\)\(^29\) as well as model calculations\(^23\)\(^–\)\(^24\),\(^30\)\(^–\)\(^31\) to determine the molecular conductance. For our illustrative purposes, here we concentrate on the model calculations, since the attention is drawn only on the qualitative behavior of the physical quantities rather than the quantitative study.

The paper is organized as follow. Following the introduction (Section 1), in Section 2, we present the model and the theoretical formulations for our calculations. Section 3 discusses the significant results, and finally, we summarize our results in Section 4.

2 Model and the synopsis of the theoretical background

Let us refer to Fig. 1. A biphenyl molecule is attached to two metallic electrodes, namely, source and drain, where each benzene ring is threaded by a magnetic flux $\phi$. In the actual experimental setup, gold (Au) electrodes are used, and, the molecule is coupled to the electrodes through thiol (SH) groups in the chemisorption technique. Here we use a simple tight-binding Hamiltonian to describe the biphenyl molecule, and within the non-interacting picture this can be expressed in this form,

$$H_M = \sum_{i} \epsilon_i c_i^\dagger c_i + \sum_{<ij>} t \left( e^{i\theta} c_i^\dagger c_j + e^{-i\theta} c_j^\dagger c_i \right)$$

where $\epsilon_i$ and $t$ represent the on-site energy and the nearest-neighbor hopping strength, respectively. $c_i^\dagger$ ($c_i$) corresponds to the creation (annihilation) operator of an electron at the site $i$. In this expression, $\theta = 2\pi \phi/N$ is the phase factor due to the flux $\phi$ (measured in units of $\phi_0 = ch/e$, the elementary flux quantum), where $N$ corresponds to the total number of carbon-type sites in each benzene ring. A similar type of tight-binding Hamiltonian is also used for the description of the electrodes, where the site energy and the nearest-neighbor hopping strength of the electrodes are represented by the parameters $\epsilon_0$ and $v$, respectively. The molecule is coupled to the electrodes by the parameters $\tau_S$ and $\tau_D$, where they (coupling parameters) correspond to the coupling strengths with the source and drain, respectively.

At much low temperatures and bias voltage, we use the Landauer conductance formula\(^32\)\(^–\)\(^33\) to calculate the conductance $g$ of the molecule which can

![Figure 1: (Color online). Schematic view of a biphenyl molecule attached to two electrodes, where each benzene molecule is threaded by a magnetic flux $\phi$.](image-url)
be expressed as,
\[ g = \frac{2e^2}{\hbar} T \]  
(2)
due to the fact that the broadening of the energy levels of the molecule due to the molecule-electrode coupling is, in general, much larger than that of the thermal broadening.\textsuperscript{32} For simplicity, we take the unit \( e = h = 1 \) in our present calculations.

3 Results and discussion

To illustrate the results, let us first mention the values of the different parameters used for the numerical calculations. In the biphenyl molecule, the nearest-neighbor hopping strength \( t \) alternates between the two values,\textsuperscript{35} those are respectively taken as \(-2.55\) and \(-2.85\). The on-site energy \( \epsilon_i \) corresponding to carbon-type sites in this molecule is set to \(-6.6\).\textsuperscript{35} On the other hand, for the side attached electrodes the on-site energy \( \epsilon_o \) and the nearest-neighbor hopping strength \( v \) are fixed to 0 and 3, respectively. The Fermi energy \( E_F \) is set to 0. Throughout the study, we focus our results for the two limiting cases depending on the strength of the molecular coupling with the source and drain. Case I: The weak-coupling limit. It is described by the condition \( \tau_{SD} \ll t \). For this regime we choose \( \tau_S = \tau_D = 0.75 \). Case II: The strong-coupling limit. This is specified by the condition \( \tau_{SD} \sim t \).

Figure 2: (Color online). \( g(E) \)-\( E \) curves for the biphenyl molecule in the weak-coupling limit, where (a) \( \phi = 0 \) and (b) \( \phi = 0.45 \).
In this particular regime, we set the values of the parameters as $\tau_S = \tau_D = 2.5$.

In Fig. 2, we display the conductance $g$ of the biphenyl molecule as a function of the energy $E$ in the limit of weak-coupling. Figure 2(a) corresponds to the conductance spectrum of the biphenyl molecule in the absence of any magnetic flux $\phi$. The conductance spectrum shows very sharp resonant peaks for some particular energy values, while for all other energies, $g$ becomes almost zero. At these resonances, $g$ approaches the value 2, and therefore, the transmission probability $T$ goes to unity since we have the relation $g = 2T$ from the Landauer conductance formula (see Eq. (2)). These conductance peaks are associated with the molecular energy levels. Thus the conductance spectrum manifests itself the energy spectrum of the molecule. In the presence of the magnetic flux, the conductance spectrum gets modified significantly. Figure 2(b) shows the $g$-$E$ characteristics for the biphenyl molecule where we set $\phi$ to 0.45. From this spectrum it is observed that the widths of the resonant peaks become much narrow compared to the case when $\phi = 0$. Not only that, some resonant peaks also disappear and get reduced height in the presence of $\phi$. Here it is examined that, the width of the resonant peaks gradually decreases as the flux $\phi$ changes from 0 to $\phi_0/2$, where $\phi_0 = eh/e$ is the elementary flux-quantum. This can be implemented as follow. In the bridge system the electron passes from the source to the drain. In this case the electron waves passing through the different paths of the molecular rings may interfere constructively or destructively, leading to stronger or weaker transmission probability across the molecule. Applying the magnetic flux, the interference condition of the electron waves among the different pathways can be controlled, and accordingly, the conductance spectrum gets modified. Thus quantum interference effect plays an important role in the study of the electron transport in a molecular bridge system. The effect of the molecular coupling is also very interesting. To emphasize it, in Fig. 3 we plot the $g$-$E$ characteristics of the biphenyl molecule in the limit of strong-coupling. Figure 3(a) represents the result for $\phi = 0$, while Fig. 3(b) corresponds to the variation when $\phi$ is set to 0.3. In this strong-coupling case, the width of the resonant peaks gets sufficient broadening compared to the weak-coupling limit. The contribution

![Figure 3](image-url)

**Figure 3:** (Color online). $g(E)$-$E$ curves for the biphenyl molecule in the strong-coupling limit, where (a) $\phi = 0$ and (b) $\phi = 0.3$.

![Figure 4](image-url)

**Figure 4:** (Color online). $I$-$V$ characteristics for the biphenyl molecule. (a) weak-coupling limit, where the red and green curves correspond to $\phi = 0$ and 0.45, respectively. (b) strong-coupling limit, where the black and blue curves correspond to $\phi = 0$ and 0.3, respectively.
of such broadening comes from the imaginary parts of the self-energies \( \Sigma_S \) and \( \Sigma_D \).\(^{32}\) It is important to note that the real parts of the self-energies provide only the shift of the energy levels. Similar to the weak-coupling case, here also the width of the resonant peaks becomes reduced with the flux \( \phi \).

Another significant feature observed from the conductance spectra is the existence of the anti-resonant state. Figure 2(a) shows that it appears across the energy \( E = -3.9 \), and for the weak-coupling limit it is also observed for the same energy (see Fig. 2(a)), but due to the smallness of the width it is not quite transparent like as in the case of strong molecular coupling. The anti-resonant states are specific to the interferometric nature of the molecule and do not appear in the conventional scattering problems like one-dimensional potential barriers, etc.

The behavior of the electron transfer through the molecule becomes much more clearly visible by investigating the current-voltage characteristics, rather than the transmission spectra. The current \( I \) is computed by the integration procedure of the transmission function \( T \) (see Eq. 1), where the function \( T \) varies exactly similar to the conductance spectra, differ only in magnitude by a factor 2, since the relation \( g = 2T \) holds from the Landauer conductance formula (Eq. 2). The variation of the current-voltage characteristics of the biphenyl molecule in the limit of weak-coupling is shown in Fig. 3(a), where the red and green curves correspond to the results for \( \phi = 0 \) and 0.45, respectively. The current exhibits staircase like structure with sharp steps as a function of the bias voltage \( V \). This is due to the existence of the sharp resonant peaks in the conductance spectrum, since the current is computed by the integration procedure of the transmission function \( T \). With the increase of the applied bias voltage \( V \), the electrochemical potentials in the electrodes cross one of the molecular energy levels and produce a jump in the \( I-V \) characteristics. The effect of \( \phi \) is significantly observed from the green curve. The current gets reduced to a large extent for this \( \phi \) value. This can be clearly understood from the conductance spectrum (see Fig. 2(b)) where the widths of the resonant peaks become much narrow compared to the case where \( \phi = 0 \). The step-like behavior almost disappears in the strong molecular coupling limit.

As illustrative example, in Fig. 4(b) we plot the \( I-V \) characteristics for the molecule in the strong-coupling limit, where the black and blue curves represent the results for the cases \( \phi = 0 \) and 0.3, respectively. The current varies almost continuously with the bias voltage \( V \) and achieves much higher amplitude than the weak-coupling limit. This phenomenon can be noticed clearly by observing the areas under the \( g-E \) curves presented in Fig. 3. Here the current amplitude is also reduced with the flux \( \phi \) (see the green curve).

To emphasize the dependence of the current \( I \) on the magnetic flux \( \phi \), in Fig. 5 we show the variation of the typical current amplitude \( I_{\text{typ}} \) as a function of the flux \( \phi \) across the molecular bridge in the limit of strong molecular coupling.

![Figure 5](image_url)

**Figure 5:** (Color online). Variation of the typical current amplitude \( I_{\text{typ}} \) as a function of the flux \( \phi \) across the molecular bridge in the limit of strong molecular coupling.

4 Concluding remarks

In conclusion, we have used a parametric approach based on the tight-binding model to investigate the electron transport properties of a biphenyl molecule attached to two metallic electrodes. In the molecule, each benzene ring is threaded by a magnetic flux \( \phi \) and the coupling of the molecule to the electrodes have been described by the Newns-Anderson chemisorption theory.\(^{22-24}\) Here we have focused our results on the aspects of (a) the molecular coupling and (b) the magnetic flux \( \phi \). Our numerical results have shown that, for a fixed molecular coupling strength, the current amplitude across
the biphenyl molecule can be regulated significantly by controlling the flux $\phi$. This aspect may be utilized in designing a tailor made nano-scale electronic circuit.

This is our first step to describe how the electron transport in a biphenyl molecule can be controlled very nicely by means of the magnetic flux $\phi$. Here we have used several realistic assumptions by ignoring the effects of the electron-electron correlation, disorder, etc. We need further study in this particular system by incorporating all these effects.

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