Symmetry-dependent spin transport properties of a single phenalenyl or pyrene molecular device

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Abstract

Spintronic systems exploit the fact that the electron current is composed of spin-up and spin-down carriers, which are more easily disturbed than electronic systems. Here, we investigate the spin transport properties of a single phenalenyl or pyrene molecule connected to zigzag graphene nanoribbon electrodes by using the non-equilibrium Green’s function formalism with density functional theory. We found the difference of the symmetry on these two molecules will bring a remarkable effect on the spin transport properties of the devices. The spin-resolved currents of the single pyrene molecular device are much lower than that of the single phenalenyl molecular device when they all connected to electrodes symmetrically. In addition, we found the change of the connected site will decrease the spin-resolved currents of the phenalenyl-based molecular device drastically, but had no longer any influence with the pyrene-based molecular device. The results will be helpful for us to further understand the transfer of the spin-carriers in the spintronic systems.

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1. Introduction

As the ever-increasing demand for computing power makes the development of traditional silicon-based semiconductor devices gradually closer to its physical limits [1]. So at this time the molecular electronics which utilize the functional electronics at the molecular level to replace the traditional silicon-based semiconductor devices came into being [2,3]. After a few decades of rapid progress, the electronic devices at single molecule scale have been able to synthesize through a variety of different bottom-up methods [4,5]. In the meantime, a series of theories that can be used to predict its transport properties more accurately have been developed [6–8]. Consequently, one can design a variety of molecular devices with the ability to replace or even beyond the performance of traditional electronic devices, such as molecular switches [9–12], molecular rectifiers [13–16], molecular field effect transistors [17–20], molecular spin filters [21–26], molecular devices with negative differential resistance (NDR) characteristics [27–33], etc. However, it is still a challenge to let the molecular device working effective as traditional silicon-based semiconductor devices [34,35]. It is because the molecular orbital characteristics and the connection type between the molecules and the electrodes, these two broad categories of main factors that influence the transport properties of molecular devices [36,37]. The twist angle between the molecules and the electrodes [38], the edge geometry [39], the connection modes [40,41], and the redox of the molecule itself [42,43] all can modulate the transport properties of molecular devices obviously. Therefore, the quantitative exploration and research on the influence of these different factors is crucial for us to understand the intrinsic transport behavior of the molecular device.

Among the above factors that can affect the transport properties of the molecular electronics devices, the symmetry of the connection sites between the molecules and the electrodes of different molecular devices and the symmetry of the molecule itself often have quite notable and similarity influences to the devices and have aroused widespread concern and research. For example, Li et al. present a fundamental investigation of the role of symmetry in the transport properties of the symmetric and asymmetric zigzag graphene nanoribbons (ZGNRs), they found that whether the ZGNRs are mirror symmetric with respect to the central plane between two edges will distinctly affect the transport behaviors of the
ZGNRs which have similar metallic band structures [44]. It has revealed the different coupling between the conducting sub-bands around the Fermi level is strongly dependent on the symmetry of the systems. Ren and Chen presented a systematic study of the effects of symmetry and stone-wales defect on the transport properties of ZGNRs [45]. It proves that the symmetry of ZGNRs makes a significant effect on its transport properties. And if we consider spin polarization, both symmetric and asymmetric ZGNRs will exhibit a semiconductor behavior which is distinct from the case of spin-unpolarized. On the basis of previous work, Kang et al. have studied the dependence of the transport properties in the zigzag silicon nanoribbons on symmetry, and the results have demonstrated that symmetric and asymmetric zigzag silicon nanoribbons have very different transport properties which have shown the same results as in ZGNRs [46].

So far, the effects of symmetry on the spin transport properties of the molecular devices have rarely been studied. Hence, in this paper, we investigate the spin transport properties of a phenalenyl molecule or a pyrene molecule connecting to the ZGNRs electrodes through two kinds of different connection mode, respectively. The results prove the difference of the symmetry on these two molecules will bring a remarkable effect on the transport properties of the devices. And the change of the connected site also plays a very important role in the spin-resolved transport behavior of the phenalenyl-based molecular devices which is consistent with previous studies [47,48].

2. Model and method

The molecular devices are illustrated in Fig. 1 where a single phenalenyl or pyrene molecule covalently bridges two ZGNR electrodes. For simplicity, the symmetrical and asymmetrical attached types for phenalenyl molecular device are named M1 and M2. The symmetrical and asymmetrical attached types for pyrene molecular device are named M3 and M4, respectively. We divide the device into three regions: left electrode, right electrode, and central scattering region artificially. The central scattering region contains a portion of the semi-infinite electrodes, thereby establishing the bonding between the molecules and the electrodes; the common Fermi level, and charge neutrality at equilibrium. The geometric optimization and spin-resolved electron transport properties were calculated by using the first-principle software package Atomistix ToolKit (ATK), which is based on density-functional theory (DFT) in combination with the non-equilibrium Green's function (NEGF) [6].

The exchange and correlations were described by the Perdew-Burke-Ernzerh of (PBE) functional under the spin-dependent generalized gradient approximation (SGGA). The wave function is expanded by the double-zeta plus polarization (DZP) basis for all atoms. The real space grid techniques are used with the energy cut off of 150 Ry as a required cut off energy in numerical integrations and the solution of Poisson equation using fast Fourier transform (FFT). The geometries of electrode and central scattering region are all optimized until average residual force on each atom is smaller than 0.05 eV Å⁻¹. The applied external magnetic fields are set to the parallel magnetic ordering for both ZGNR electrodes and central molecule. The spin-resolved current \( I\left(\alpha, \beta\right) \) of the junctions can be calculated by the spin-resolved Landauer formula:

\[
I\left(\alpha, \beta\right) = \frac{e}{h} \int T\left(E, V_b\right) f_{L}(E) \left[ f_{R}(E, V_b) - f_{R}(E, V_b) \right] dE
\]

Here, \( T\left(E, V_b\right) \) is the spin-dependent transmission coefficient, \( f_{L}(E) \) and \( f_{R}(E, V_b) \) are the Fermi-Dirac distribution functions of the left and right electrodes, and \( \sigma \) represents the \( \sigma \)-spin and \( \beta \)-spin state. The total transmission probability is

\[
T\left(E\right) = T_{\sigma}(E) \left| G_{\sigma}(E) \right|^2 \left| G_{\sigma}(E) \right|^2
\]

where \( G_{\sigma}(E) \) and \( G_{\sigma}(E) \) are the retarded and advanced Green functions of the central region. Therefore, \( I\left(\alpha, \beta\right) \) for electrons incident at an energy \( E \) through the device under the potential bias \( V_b \) is composed of all available conduction channels with the individual transmission \( T_{\sigma} \).

3. Results and discussion

First, the spin-resolved current-voltage (I-V) characteristics of device M1-M4 under finite bias voltage are shown in Fig. 2. One can clearly observe that the symmetry of the molecule plays an important role in the spin-resolved transport properties. In Fig. 2(a), when a phenalenyl molecule covalently bridges two ZGNR electrodes at the same connect site, the \( \alpha \)-spin currents and \( \beta \)-spin currents perform a linearly increasing with the applied bias voltage. The values of two spin transport currents are almost identical in the whole bias region from \(-0.8 \) V to \(0.8 \) V. In addition, their I-V characteristics are symmetrical in positive and negative bias region due to the vertical symmetrical geometry of the device. In Fig. 2(b), after the right attached site on phenalenyl molecule changes, the forward symmetrical geometry of M1 is broken in M2 because the phenalenyl is only a vertical symmetrical molecule. We found the spin-resolved currents of M2 are obviously decreased in comparison with that of M1 and the maximal spin-resolved currents do not exceed 1.5 \( \mu \)A. In addition, the \( \alpha \)-spin currents will increase faster than \( \beta \)-spin currents in the positive bias region. Nevertheless, it is just the opposite under the negative bias, the \( \beta \)-spin currents increase faster than the \( \alpha \)-spin currents. Therefore, the I-V characteristics of \( \alpha \)-spin current and \( \beta \)-spin current are asymmetrical in positive and negative bias region.

For M3, when a pyrene which is a vertical and horizontal symmetrical molecule bridges two ZGNR electrodes at the same site covalently, the \( \alpha \)-spin and \( \beta \)-spin currents are also identical in the whole bias region (see Fig. 2(c)). The symmetrical I-V characteristics also can be observed in positive and negative bias region due to the vertical symmetrical geometry. However, the values of the spin-resolved currents are lower than that of M1 obviously. The maximal spin-resolved currents of M3 do not exceed 0.06 \( \mu \)A. In Fig. 2(d), after the right attached site on pyrene molecule changes, the values of the spin-resolved current of M3 don’t show much difference in comparison with that of M3. The maximal spin-resolved currents also do not exceed 0.06 \( \mu \)A. More importantly, the asymmetrical I-V characteristics are not so as M2 due the vertical and horizontal symmetrical geometry of the pyrene molecule.

By comparing the spin-resolved transmission spectra and the spin-resolved spatial distributions of local density of states (LDOS) at Fermi level, we are able to analyze the influences of the molecular symmetry on their spin-resolved transport properties. In Fig. 3(a), one can see the \( \alpha \)-spin and \( \beta \)-spin transmission spectra are vertical symmetrical and there are a series of strong transmission peaks nearby the Fermi level. That’s why two kinds of spin currents of device M1 perform a good conduction even at a lower bias voltage and always show the symmetrical I-V characteristics in Fig. 2(a). Because the phenalenyl is just a vertical symmetrical molecule, the changing of the connected site in M2 will break the symmetry of the device. So, the \( \alpha \)-spin and \( \beta \)-spin transmission spectra of M2 are no longer same to that of M1 in Fig. 3(a). In addition, one can see the number of both the \( \alpha \)-spin and \( \beta \)-spin transmission peaks around the Fermi level decreases correspondingly and their transmission coefficients are significantly reduced in compassion with M1. So, the transport performance of M2 at low bias voltage will be greatly weakened in compassion with M1.

In order to understand how the change of the connection site between molecule and electrode affects the transport spectra of M1 and M2, we illustrate their spin-resolved spatial distributions of LDOS at Fermi level in Fig. 3(c) and (d). LDOS is a physical quantity that describes the space-resolved density of states (DOS), which is
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