Carbon 110 (2016) 200–206

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

Giant decreasing of spin current in a single molecular junction with twisted zigzag graphene nanoribbon electrodes

Zhi-Qiang Fan [a, b], Fang Xie [c], Xiang-Wei Jiang [b, *], Zhongming Wei [b], Shu-Shen Li [b]

[a] School of Physics and Electronic Science, Changsha University of Science and Technology, Changsha, 410114, People's Republic of China
[b] State Key Laboratory for Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing, 100083, People's Republic of China
[c] Physics Science and Engineering Technology College, Yichun University, Yichun, 336000, People's Republic of China

ARTICLE INFO

Article history:
Received 4 July 2016
Received in revised form
23 August 2016
Accepted 9 September 2016
Available online 9 September 2016

ABSTRACT

Molecular spintronics is a new and emergent sub-area of spintronics that has the potential use in future information storage, magnetic sensing and quantum computing. We investigate the spin transport properties of a single benzene molecule connected to zigzag graphene nanoribbons (ZGNRs) by using a self-consistent *ab initio* approach which combines the non-equilibrium Green's function (NEGF) formalism with density functional theory (DFT). The spin-resolved current-voltage characteristics of the single benzene molecule at finite biases are different while the left and right zigzag graphene electrodes with the parallel (P) and anti-parallel (AP) magnetism configurations. The perfect (100%) spin polarization in a large bias region can be realized with both P and AP magnetism configuration. However, the spin-resolved rectifications are only found with AP magnetism configuration. More importantly, both of the α-spin and β-spin currents would drop remarkably when one ZGNR electrode is twisted. Especially the α-spin currents with P magnetism configuration will decrease by up to 8 orders of magnitude when the twisted angle reaches 90°. The above results demonstrate that this junction holds promise in the design of a high-performance multifunctional single-molecule spintronic device.

1. Introduction

Over the past few decades, minimization of the device size is an important challenge for near-future electronics to realize low-cost device with low power consumption and high functionalities. Fundamental studies on molecular-scale electronics have rapidly developed in recent years [1–4]. Among them, graphene and graphene based molecular-scale devices have drawn great attention owing to their remarkable physical properties and application potential in future nanotechnology [5–10]. The advanced fabrication and patterning technology demonstrated that graphene can be cut into many different shapes and sizes, such as 1D graphene nanoribbons (GNRs) [11,12], graphene quantum dots [13], graphene quantum rings (GQRs) [14], and graphene nanoflakes (GNFs) etc. [15,16]. The various nonlinear current-voltage characteristics, such as negative differential resistance (NDR) [17–20], conductance switching [21–25], and electrostatic rectification [26–29] have been found in above graphene based junctions. These interesting and important nonlinear properties have profound potential for future molecule-scale circuits.

The spin magnetism is another important property of graphene which have attracted much attentions in recent years [30,31]. Generally, zigzag edged GNRs (ZGNRs) were predicted to be half-metallic under an electrical field. Kan et al. presented a detailed study on the electronic properties of several edge-modified ZGNRs and found the half-metallicity can be realized in ZGNRs by edge modification [32]. Kang et al. had investigated the magnetic and spin dependent transport properties of asymmetrically hydrogenated ZGNRs and demonstrated that it is possible to realize perfect spin filtering effect on H2–ZGNR–H by p-type or n-type doping [33]. Soon, Zeng et al. investigated the electronic transport properties of the mono-hydrogen terminated and dihydrogen-terminated ZGNR hetero-junctions [34]. Results show that a perfect spin-filtering effect with 100% spin polarization can be realized by dehydrogenation. Most recently, Cui et al. investigated the magnetism and spin transport properties of hetero-structures based on zigzag graphene nanoribbons with edge hydrogenation and oxidation [35]. An excellent dual spin filtering behavior was

* Corresponding author.
E-mail address: xwjiang@semi.ac.cn (X.-W. Jiang).

http://dx.doi.org/10.1016/j.carbon.2016.09.022
0008-6223/

obtained from their hetero-structures. Beside, the spin transport properties of the single molecules connected to ZGNRs have also drawn great attention. The perfect spin-valve and spin-filter effects had been found in ZGNR-carbon wire junctions in 2010 [36]. Soon, Zhang et al. found the poly-(terphenylene-butadiynylene)s molecule can perform the high bipolar spin filtering and switching behaviors between ZGNR leads [37]. Then, Li et al. calculated spin-resolved current-voltage character of an oligo p-phenyl-enevinylene molecule between two ZGNR leads and found the excellent NDR and spin-rectifying behaviors [38]. The above results open the possibility of spintronics application for graphene.

In this paper, we use the first-principles approach based on the DFT combined with the NEGF technique to investigate the spin transport properties of a single benzene molecule connected to ZGNR electrodes. The aim is to reveal why the phenyl-based molecules will show different spin-resolved current-voltage characteristics when the connected ZGNR electrodes with the P and AP magnetism configurations [39,40]. In addition, the previous first-principles quantum calculations show that the thermal conductance of the orthogonal junction does not change very much at low temperatures when one connected ZGNR electrode is rotated by 90° [10]. However, some previous works show the twist in graphene can modulate its electronic transport properties effectively [41–45]. For example, twisting can tune the band gap of H-terminated or F-terminated armchair graphene nanoribbons (AGNRs), and even close the band gap under certain twist angle [43,44]. Nabil et al. predict the conductance of ferromagnetic ZGNR depend on the twisting angle remarkably, and suggest that a spin valve function in a flexible nanoribbon can be realized by twisting in ferromagnetic ZGNRs [45]. Inspired by these work, we also investigate the effect of ZGNR electrode twisting on the spin-resolved current-voltage characteristics of the benzene molecule. The results show that both of the α-spin and β-spin currents would drop remarkably when the right ZGNR electrode is twisted. The systematic analyses on spin densities, transmission spectra, transmission pathways, and the projected density of states (PDOS) explore the physical mechanisms of the above results.

2. Model and method

The molecular device is illustrated in Fig. 1(a). A single benzene molecule covalently bridges two ZGNR electrodes consisting of 4 zigzag carbon chains. The device is divided into three regions: left electrode, right electrode, and central scattering region. The central scattering region contains three units of ZGNR electrode on each side, thereby establishing the bonding between the molecules and the electrodes, the common Fermi level, and charge neutrality at equilibrium. In Fig. 1(b), we fix the left ZGNR electrode and only twist the right ZGNR electrode around the central carbon wires to investigate the effect of one ZGNR electrode twisting on the current-voltage characteristics of the junctions. The θ is the twisted angle between the left ZGNR electrode and right ZGNR electrode. For simplicity, four junctions (θ = 0°, 30°, 60°, and 90°) are considered in this work which are named M1, M2, M3 and M4, respectively. Two types of magnetic configurations, parallel (P) and anti-parallel (AP), for electrodes are considered here, i.e., the external magnetic fields applied on two electrodes are set to point to the same or opposite direction, thus exhibiting the parallel [↑, ↑] or anti-parallel [↑, ↓] magnetic ordering for both ZGNR electrodes. Isosurface plots of the spin charge density difference between α-spin and β-spin states ($\nabla\rho = \rho_\alpha - \rho_\beta$) for M1 with the P and AP magnetism configurations are shown in Fig. 1(c). Here, ρ_α and ρ_β denote the electron density of α-spin (red) and β-spin (blue), respectively.

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) [46,47]. The exchange-correlation potential is described by the local spin density approximation (LSDA), which works rather well for light elements and systems where electrons are delocalized. The wave function is expanded by a single-zeta plus polarization (SZP) basis for H atoms, and double-zeta plus polarization (DZP) basis for C atoms. The k-point sampling is 1, 1, and 100 in the x, y, and z directions, respectively, where the z is the period direction of nanoribbons. 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 are optimized until all residual force on each atom is smaller than 0.05 eV/Å. The spin-resolved current $I_{(\alpha,\ \beta)}$ of the junctions can be calculated by the spin-resolved Landauer-like formula: $I_\sigma(V_b) = \frac{e}{h}\int T_\sigma(E,V_b)[f_L(E,V_b) - f_R(E,V_b)]dE$ [48,49]. Here, $T_\sigma(E,V_b)$ is the spin-dependent transmission coefficient, $f_L(E,V_b)$ and $f_R(E,V_b)$ are the Fermi–Dirac distribution functions of the left and right electrodes, and σ represents the α-spin and β-spin state. The transport coefficient $T_\sigma(E,V_b)$ can be calculated using the well-known formula, $T_\sigma(E,V_b) = Tr[\Gamma_L(E)G^R(E)\Gamma_R(E)G^A(E)]$, where $G^R(E)$ and $G^A(E)$ are the retarded and advanced Green's functions, respectively. $\Gamma_{L,R}$ are the coupling functions of the conductor to the left and right electrodes.

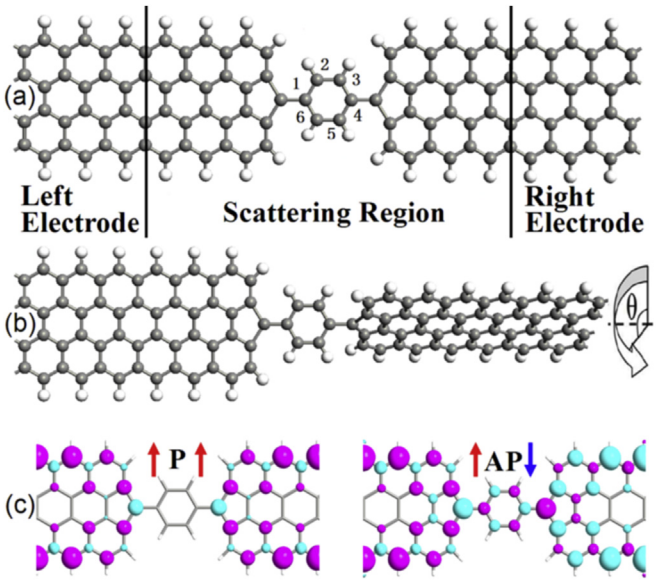

Fig. 1. (a) and (b) Schematic view of an atomic carbon wire-graphene junction. All edge carbon atoms in GNR are saturated with hydrogen atoms. The leftmost carbon atom in the benzene molecule is labeled as C1 and other five carbon atoms are labeled as C2, C3, C4, C5, and C6 in clockwise direction. The θ is the twisted angle of the right ZGNR electrode relative to the left ZGNR electrode. (c) Isosurface plots of the spin charge density difference of α-spin and β-spin states for M1 with the P (left) and AP (right) magnetism configurations. Isovalue is fixed at 0.5 for two magnetism configurations. (A colour version of this figure can be viewed online.)

3. Results and discussion

The calculated spin-dependent transmission spectra for M1, M2, M3 and M4 are presented in Fig. 2(a)-(h). As the previous relative articles suggest, the spin transport properties of our junctions are also different when the connected ZGNR electrodes with the P and AP magnetism configurations. In Fig. 2(a), the transmission coefficient of α-spin state is much larger than that of the β-spin state in