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The effect of asymmetrical electrode on the transport properties of molecular devices

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ABSTRACT

By applying nonequilibrium Green's functions in combination with the density functional theory, we have investigated the electronic transport properties of molecular devices consisting of the carbon atomic chain coupling with symmetry and asymmetry Au electrodes. The asymmetry Au electrodes systems display good rectifying behavior. The main origin of this phenomenon is that a molecular core coupling with asymmetry electrodes can generate two asymmetrical Schottky barriers at both extended molecule regions. This rectification is also explained by the calculated transmission spectrum and the spatial distribution of the LUMO and HOMO states.

1. Introduction

The molecular devices have been attracting much attention owing to their underlying physics and potential practical application. Many interesting transport properties, including molecular rectification [1–5], NDR [6–11], and highly nonlinear current–voltage relationship [12–16], were found in many molecular devices. Especially the molecular rectifier, proposed first by Aviram and Ratner [17], will play a key role for the future development of molecular electronics because it is the simplest functional element for building an electronic circuit [18]. Generally speaking, the origins of the molecular rectification in a molecular device may have several important causes as follows: (1) the asymmetrical electrode materials generating two asymmetrical Schottky barriers at contacts [19], (2) the matching degree of the orbitals around the Fermi level among the two electrodes and the molecule [20], and (3) the asymmetric shift and different alterations of the frontier molecular orbitals peaks under bias of different polarities [21]. Among previous studies, the sensitivity of transport properties of a molecular device to electrode materials or contact groups has been investigated extensively. But these studies have little consideration on the effect of electrode structures.

Carbon-based systems, such as carbon atomic chains [22–32] and its derivatives, are another class of materials which have gained more attention. Linear carbon atomic chains have also been realized by removing carbon atoms row by row from graphene through the controlled energetic electron irradiation inside a transmission electron microscope [33]. When it is embedded between two electrodes, the system may display the rectifying behavior and NDR effect [34]. In this paper, by using first-principle calculations, we report the results of electronic transport properties of carbon atomic chains connected to symmetry and asymmetry Au electrodes. We find that rectifying behaviors are observed in the asymmetry Au electrodes systems.

2. Model and method

The transport properties of molecular devices are calculated based on nonequilibrium Green's function formalism as implemented in Atomistix Tool Kit (ATK), which is based on real-space, nonequilibrium Green's function formalism and the density-functional theory. In our calculations, the local density approximation (LDA) schemes are used with the Perdew–Zunger parametrization of the correlation energy of a homogeneous electron gas calculated by Ceperley–Alder. The electrode calculations are performed under periodical boundary conditions, and the *k*-point grid being $1 \times 1 \times 100$. All configurations are relaxed until their force tolerance being less than 0.05 eV/Å.

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Self-consistent calculations are performed with a mixing rate set to be 0.02, and the mesh cutoff of carbon atom is 150 Ry to achieve a balance between calculation efficiency and accuracy.

The molecular devices we study are illustrated in Fig. 1. The system is divided into three regions: left electrode, scattering region, and right electrode. The carbon atomic chain (C_7 and C_8) is coupled to two one-dimensional semi-infinite Au electrodes, and

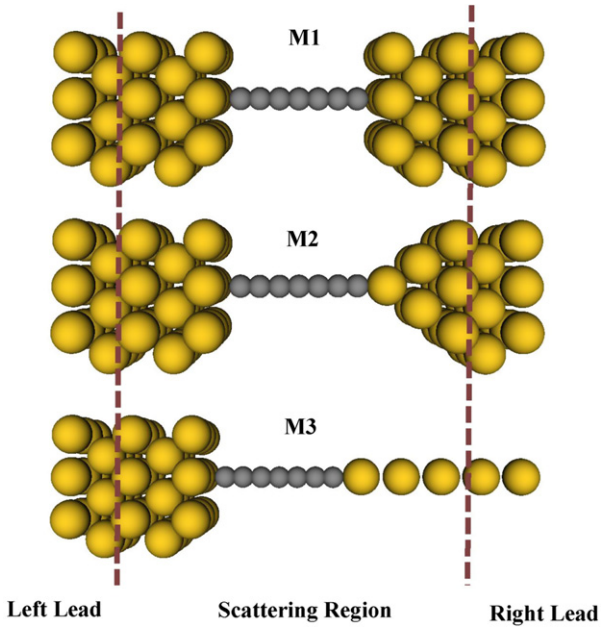

Fig. 1. Structures of molecular devices in our simulation. A carbon atomic chain is sandwiched between left electrode (L—Au (1 1 1)) and right electrode (R—Au (1 1 1), Au pyramid, Au atomic chain), which is referred to as models M1, M2, and M3.

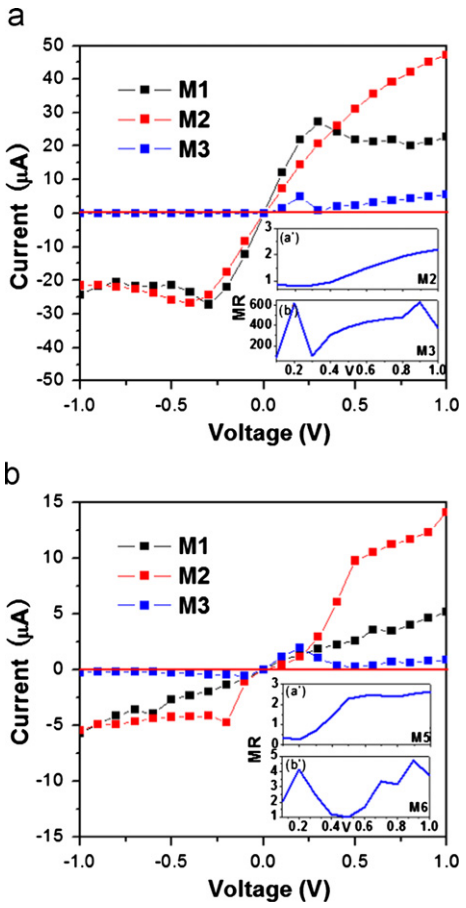

Fig. 3. Calculated *I–V* curves for central molecule C_7 chain (a) and C_8 chain (b) in M1–M3 models. The insets (a)' and (b)' show the rectifying ratio for M2 and M3, respectively.

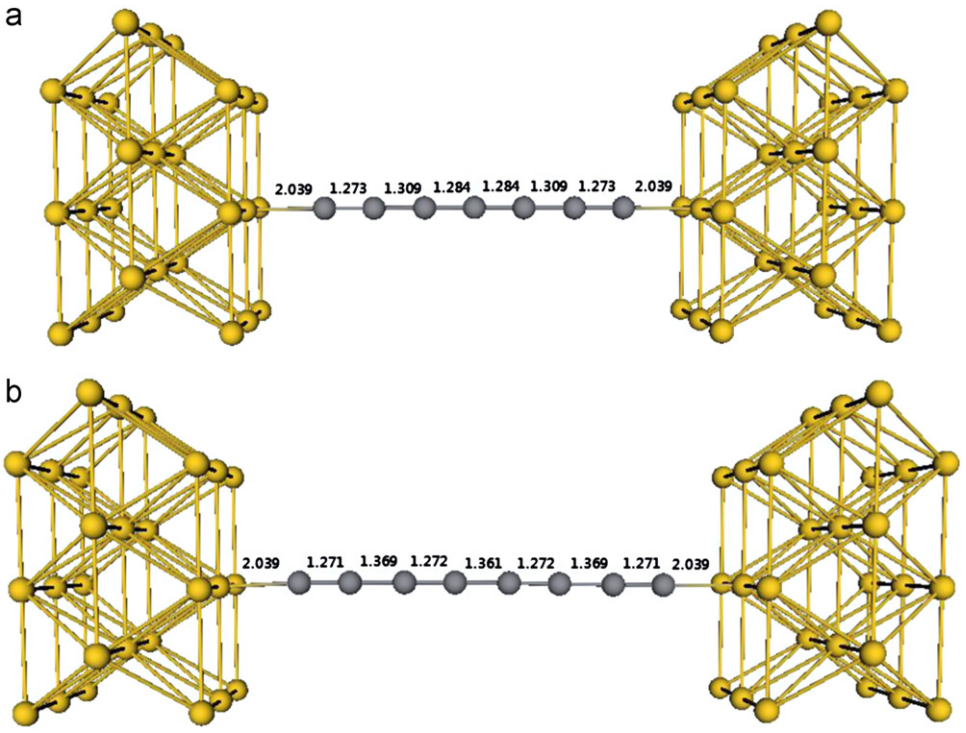

Fig. 2. Bond length of C_7 (a) and C_8 (b) after the model optimizing.