Quantum transport of molecules and internal current

Masaru Tsukada
Dept. of NanoSci. and NanoEngin.,
Waseda University

二つのナノ電極を結ぶ原子細線、および分子架橋系の量子伝導の特徴を議論した。原子細線については、密度汎関数法（DFT）の非平衡開放系への拡張であるリカージョン伝達行列法（RTM）を、分子架橋系については非平衡グリーン関数法によって解析した。分子架橋の縮重準位付近の共鳴トンネルで現れる分子内の大きなループ電流と永久電流との関係など、興味深い性質を論じた。

Electron transport through atomic wires and molecular bridges connecting nano-scale electrodes has attracted much attention recently. Because of the difficulties in the fabrication, the role of theoretical studies are very significant for developing novel quantum functions and providing guiding principles for promising molecular and other nano-structures. We discussed some remarkable properties of quantum transport through atomic wires and molecular bridges based mainly on theoretical results from the author's group.[1)]

The first topic is the quantum transport properties of atomic wires revealed by the recursion transfer matrix method (RTM)[2)], which is an extension of the conventional density functional method (DFT) to non-equilibrium open systems. Recently, we succeeded to implement non-local pseudo-potentials to the RTM[3)]

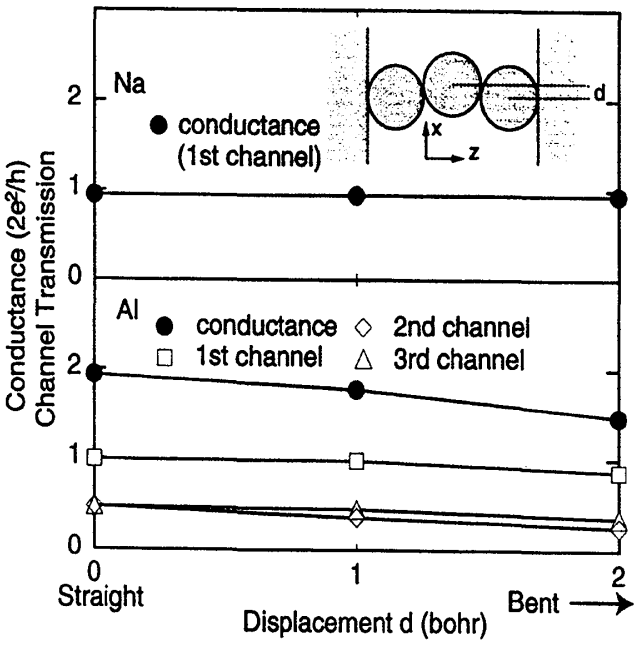
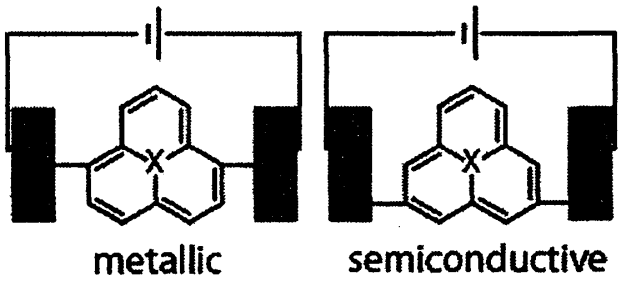


Fig.1
Conductance of Na and Al 3 atom atomic bridge as the function of bending displacement

method. Based on the calculation, physical quantities such as conductance, the density of states (DOS), and the transmission coefficients are decomposed into the eigen-channel components.

Figure 1 shows the conductance and the channel transmission of Al wires as a function of a displacement d of the central atom. For the comparison, we also show the conductance of Na wire in the same figure. For the Al wire, three channels contribute to the conductance: one almost fully open channel and two half open channels. With increasing d, each channel transmission decreases, thus the conductance also decreases for the Al wire. For the bent wires, a loop current is found circulating around the corner atom. The position of the onset energy of each channel as well as the dominantly contributing atomic orbitals, is important for understanding the behavior of channels. We also investigated the conductance through Al atomic wires composed of six atoms with a different kind atom (from Na to Cl) connecting to the left electrode. The conductance is strongly dependent on the bonding nature of the atom at the contact.

For a theoretical approach to the transport of molecular



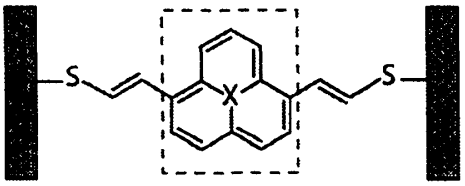


Fig.2 Molecular bridges of Phenalenyl

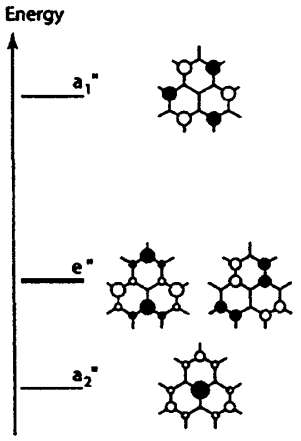


Fig.3
Molecular orbitals of phenalenyl

bridges, we use the non-equilibrium Green's function method (NEGF) with tight binding basis.[4] The quantum transport through molecular bridges is strongly influenced by the connecting part to the electrodes. The case study is done for the phenalenyl molecule.[5] The phenalenyl-like molecule are attached to the two semi-infinite gold electrodes through the mercapto-vinyl groups as seen in Fig.2. The transmission spectra when both leads are

attached to the α site or β site shows metallic or semiconductor feature, repsctively. The dependence on the connecting sites of leads is explained by the wavefunction near the Fermi level shown in Fig.3.

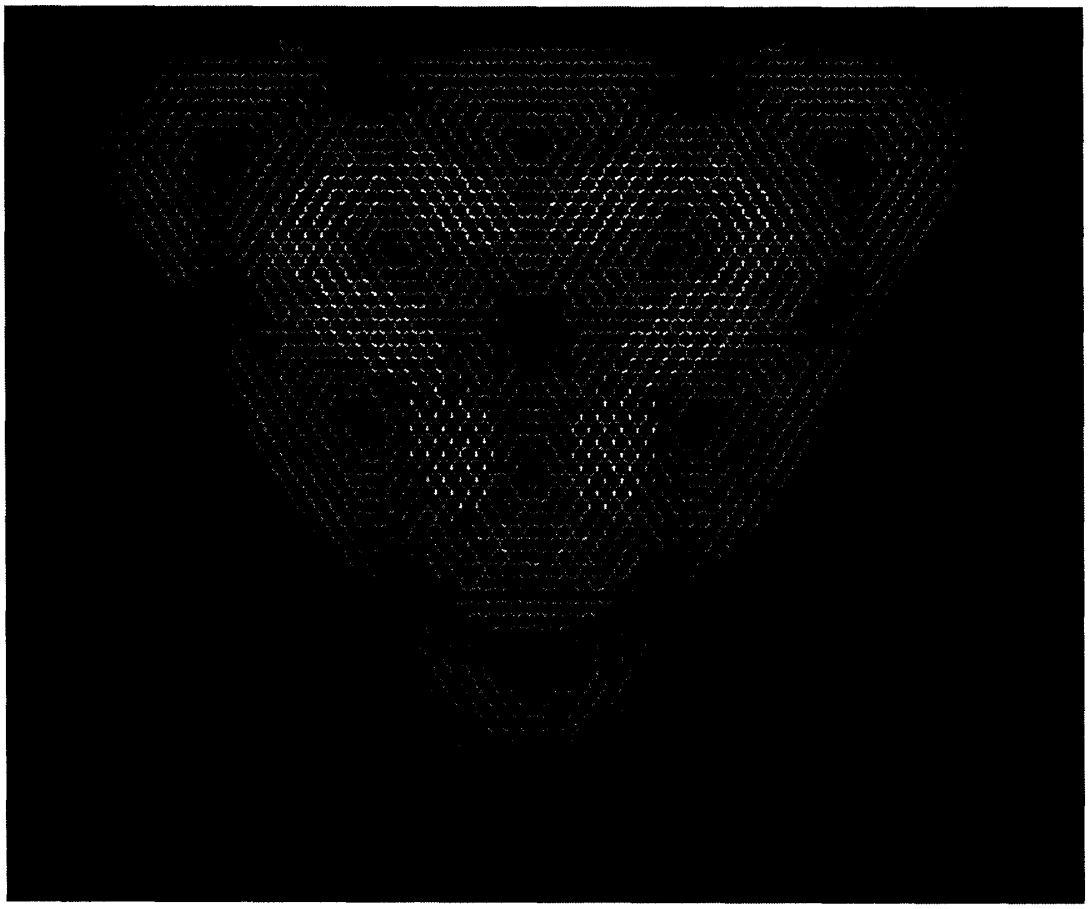


Fig.4
Large loop current of triangular monolayer graphite induced by source-draine current

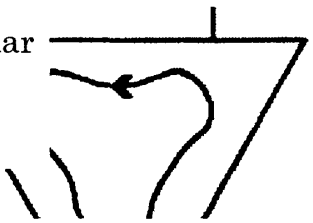


Fig.5
Schematic view of the loop current

Figure 4 illustrates a triangular nano-graphene sheet bound with zigzag edges. In this model, the source and drain leads are modeled by semi-infinitely long non-interacting atomic chains.The protruded atoms along the topmost zigzag edge are numbered from 1 to N. In total the number of atoms in the nano-graphene is $N^2 + 4N + 1$. In the following, we focus on the large system with N = 56.[6] The source and drain leads are connected to

the nano-graphene molecule at the sites (s,d) = ($N/4$ +1, 3 N /4). The isolated nano-graphene molecule has many doubly degenerate energy levels in the vicinity of the Fermi level. Although there are (N-1)-fold degenerate levels at E = 0 corresponding to the edge states, they do not contribute to the resonant current. Figure 4 shows the internal bond current $J_{ji}(E)$ (from site i to j) at one of the degenerate levels E=-0.165 t, so that the strongest one is expressed by the darkest color.

Figure 5 schematically illustrates the current flow. Noticeably, the current does not circulate around an individual small Kekul'e unit, rather winds inside the nano-graphene circulating a large area. The strength of the current is much larger than the source-drain current. Even when the source and drain sites are moved to different sites along the topmost side of the nano-graphene, almost the same current patterns are obtained at these degenerate energy levels. It suggests that the internal current distribution is determined from the nature of the particular degenerate molecular orbitals, rather than being accidentally induced by the current from the leads.

As a general feature, when the electron incident energy is close to a degenerate molecular level, a strong loop current is generated inside the molecule. For larger molecules, the internal current distribution is similar to that induced by the magnetic field, for example the persistent current. Usually the orbital diamagnetism of a molecule is contributed by an incomplete cancellation of the magnetic moment contributed from internal currents of opposite chiralities. Current distribution of either chirality shows almost the same feature as the source-drain induced loop current.

Refernces

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