論文 / 著書情報
Article / Book Information

題目(和文)	
Title(English)	Study of Electron Transport through Rigid and Planar Single Molecular Wires
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出典(和文)	学位:博士(工学), 学位授与機関:東京工業大学, 報告番号:甲第10919号, 授与年月日:2018年6月30日, 学位の種別:課程博士, 審査員:真島 豊,金 有洙,伊藤 満,梶原 正憲,多田 朋史,東 康男
Citation(English)	Degree:Doctor (Engineering), Conferring organization: Tokyo Institute of Technology, Report number:甲第10919号, Conferred date:2018/6/30, Degree Type:Course doctor, Examiner:,,,,,
学位種別(和文)	博士論文
Category(English)	Doctoral Thesis
種別(和文)	要約
Type(English)	Outline

Thesis Outline

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In this thesis, I have characterized single molecular wires by using both electroless Au-plated Au/Pt-based nanogap electrodes and scanning tunneling microscopy (STM) for demonstration stable coherent resonant tunneling and Coulomb blockade, and also single molecular wire resonant tunneling transistors.

I have characterized electrons transport through COPV6$(SH)_2$ molecular wires introduced between nanogap electrodes. Two types of devices are observed as SAuSH device that one-side sulfur terminal is chemically bonded onto electrode, and $(SH)_2$ device that molecular wire is physically absorbed between nanogap electrodes. Based on two different structures, I demonstrated two different electron transportat mechanisms, one is coherent resonant tunneling on SAuSH device, and the other is sequential single-electron tunneling on $(SH)_2$ device (Chapter 2). I observed current-voltage characteristics from SAuSH device of COPV5$(SH)_2$ molecular wires and one dI/dV peak is largely shifted under the application of gate bias voltage. This gate oscillation suggests coherent resonant tunneling modulation through molecular orbitals. (Chapter 3). On a weakly coupled SAuSH device of COPV5$(SH)_2$ molecular wires, I observed current limited bistability behavior at 284 mK (Chapter 4). This bistablity behavior is discussed by change in molecular wire orientation due to adding/removing one electron from molecular wire. Additional experimental results by using STM are discussed in chapter 5 for comparison. I introduced COPV2 molecular wire on Ag(111) and NaCl/Ag(111) surfaces by thermal evaporation. I investigated single long molecular wire (COPV6) one-side chemically absorbed on Ag(111) surface. Those two results are important compensation for this thesis to understand electron transportation mechanism in real space.

I summarized this thesis by every chapters as follows:

Chapter 2: Coherent Resonant Tunneling and Sequential Single Electron Tunneling through COPV6(SH)2

The fabrication processes of stable nanometer scale electrodes are introduced in detail by using electron beam lithography and electroless Au-plating (ELGP) process. Electroless Au-plated Au-based nanogap electrodes are used. COPV6 long, rigid and planar molecular wire was introduced between the nanogap electrodes. Three structures of COPV6 devices are expected as $(SAu)_2$, SAuSH, and $(SH)_2$ devices. From all of 100 devices, no $(SAu)_2$ device can be found. The results are divided into two categories, one is large no current voltage region followed by current level of 3 nA at high drain bias of 1.5 V. Here, coherent resonant tunneling through molecular orbitals (HOMO and LUMO) is observed for this SAuSH device, and confirmed by DFT calculation of energy levels spacing. This coherent resonant tunneling phenomena were also observed even at room temperature. The other is small no current voltage region followed by low current level less than 100 pA at drain bias of 0.8 V. Sequential single-electron tunneling through molecular wire on $(SH)_2$ device is confirmed by orthodox model of single-electron effect., in which COPV6 is physisorbed between nanogap electrodes.

Chapter 3: Single Molecular Resonant Tunneling Transistor

Single SAuSH device was fabricated by using electroless Au-plated Pt-based nanogap electrodes and COPV5$(SH)_2$ molecular wire, as we mentioned in last the chapter. The smaller line widths of source and drain electrodes are prepared, which enables us to increase gate capacitance. Three-terminal system is formed by source, drain and two side gate electrodes. Single molecular resonant tunneling transistor operation is obtained. This gate oscillation suggests coherent resonant tunneling modulation through molecular orbitals.. We explained this behavior by introducing molecular radical states. Electrons coherent resonant tunnel through molecular wire orbitals when a molecular orbital energy level aligned with the Fermi energy of the electrodes.

Chapter 4: Current Limited Bistability in COPV5$(SH)_2$ Nanodevice

COPV5$(SH)_2$ molecular wire was introduced between nanogap electrodes by immersing substrate into 0.5 mM solution for 3 h at 80 ℃ for spontaneously absorption. The substrate was stabilized onto package, and was kept in cryogenic temperature of 284 mK by using 3He refrigerators with sorption pump. I found that bistability behavior was observed at onset of current flow, and current level is limited at around 200 pA and -200 pA for both positive and negative bias, respectively. Current limited bistability was observed in COPV5 nanodevice, which was explained as change in molecular wire orientation due to adding/removing one electron from molecular wire. Then, the frequency of observed bistability was discussed at Poisson Process, showing random telegraphy signal. Possible mechanism was given as atomic scale mechanic movement for molecular wire weakly coupling to electrodes.

Chapter 5: COPVn Molecular Wire on Ag(111) Investigated by STM

A single molecule was deposited onto Ag(111) surface, the tip/vacuum/molecule/metal system is similar to single molecule between nanogap electrodes. A COPV2 molecular wire was directly evaporated onto Ag(111) surface at 180 ℃ for 3 min by a home-made evaporator. According to the orientation of molecular wires, the shape of molecules on STM images can be concluded into two types (dark and bright in center). Both of them indicate that molecules were decoupled from metal surface by their side chains.