CONDUCTANCE SWITCHING IN SINGLE REDOX MOLECULES

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The building blocks of silicon-based microelectronics are field effect transistors (FET) whose basic function is to switch electrical current between two electrodes on and off with a third electrode (gate). Building a single molecule FET is naturally considered to be a critical step towards the ultimate goal of molecular electronics. FET-like behavior has been demonstrated in carbon nanotubes[1] and semiconductor nanowires,[2] and Coulomb blockade[3-6] and Kondo effects[4-5] have been recently observed in molecular systems using a back gate at low temperatures. Theoretical models have predicted that the conductance of a single molecule can be modulated with a gate electrode via resonant tunneling[7-8]. Experimental demonstration of this FET behavior in single molecules has, however, been a difficult challenge because it requires one to: 1) find a reliable method to wire a single molecule to the source and drain electrodes; and 2) place the gate electrode a few Å away from the molecule to achieve the required gate field.

We have demonstrated single molecule field effect transistors (FET) which consists of a redox molecule covalently bonded to a source and drain electrode and an electrochemical gate (Fig. 1). By adjusting the gate voltage, the energy levels of empty molecular states are shifted to the Fermi level of the source and drain electrodes. This results in a large increase in the source-drain current, in a fashion of FET. The large current increase is attributed to an electron transport mediated by the lowest empty molecular energy level when it lines up with the Fermi level. This behavior has been observed in several redox systems.

One is perylene tetracarboxylic diimide (PTCDI) terminated with thiol groups has been studied with this method. Based on the statistical analysis, the average conductance of single PTCDI has been found at 1.2×10^{-5} G_0 (Fig. 2). Large gate effect has been discovered on PTCDI, which resembles the n-type solid state FET. With fixed source-drain bias, the current flow through single PTCDI molecule can be reversibly controlled 3 orders at room temperature by electrochemical gate. The possible mechanism for the large gate effect of PTCDI is resonant tunneling or two-step electron transport. In contrast, oligothiopehenes terminated with thiol groups (with 3 and 4 repeating units) exhibit p-type transistor characteristics.

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Figures:

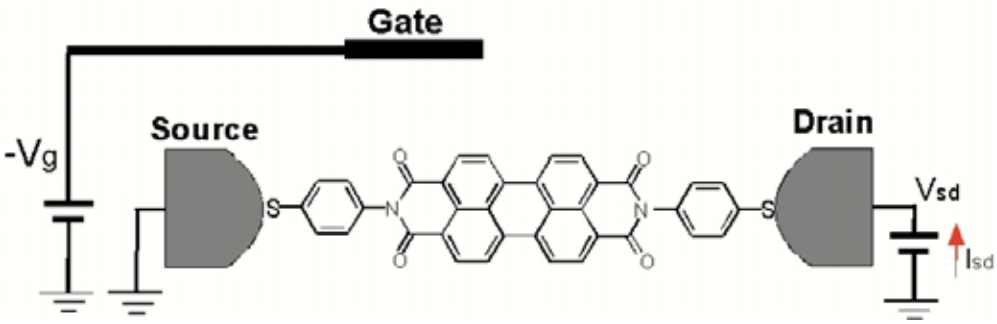

Fig. 1 Schematic of a single molecule transistor with an electrochemical gate. The gate and the source-drain bias voltages are controlled with a bipotentiostat.

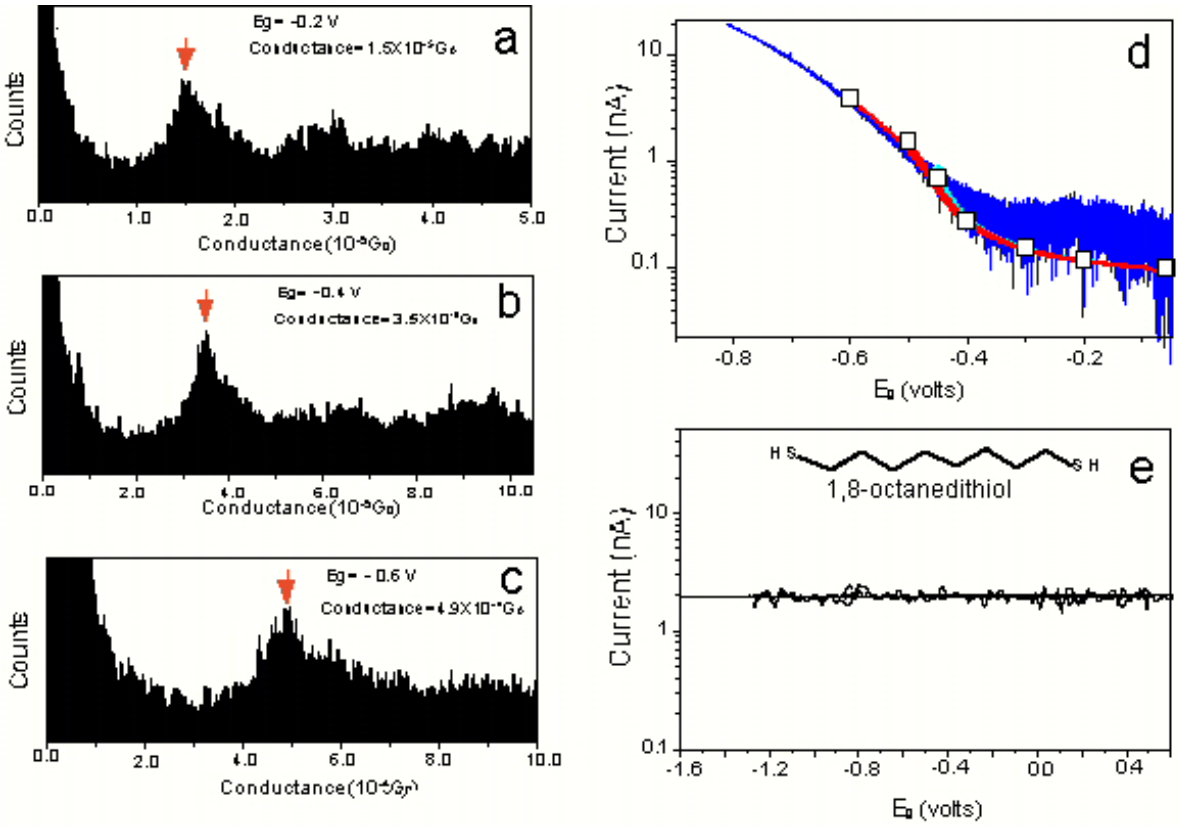

Fig. 2 (a-c) Conductance histogram constructed from ~1000 individual transient curves at different gate voltages. (d) Source-drain current (I_{sd}) vs. gate voltage (V_g) for a single PTCDI molecule transistor. The open squares were obtained from the peak position of the conductance histograms. The solid lines were obtained by directly recording the source-drain current while sweeping the gate voltage. (e) Control experiment on an alkanedithiol shows no gate-voltage dependence.

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