Electronic Supplementary Information

Electronic transport in Benzodifuran single-molecule transistors

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(Dated: March 27, 2015)

Contents

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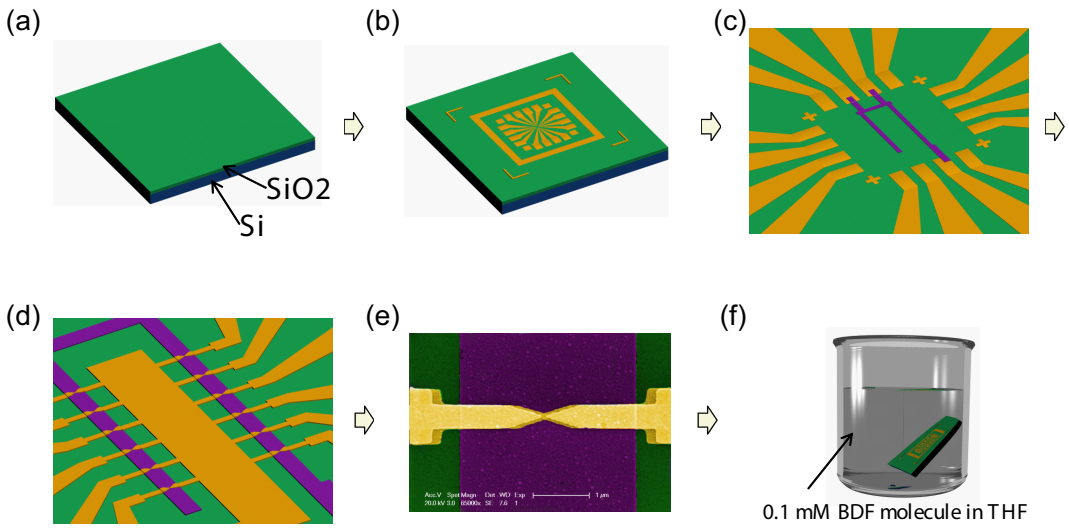

FIG. S1: **Fabrication procedure of BDF single-molecule transistors.** **(a)** A Si/SiO_2 wafer as a substrate. (b) bonding pads and interconnects defined photolithographically. **(c)** Deposition of Al film (purple area) covered with a thin layer of naturally oxide (AlO_x) for the gate electrode. **(d)** Fabrication of gold constrictions with the thickness of ~20 nm on the AlO_x layer by electron beam lithography. **(e)** False color scanning electron microscopy image of an as-fabricated device. **(f)** The devices were immersed in the dilute BDF solution overnight to form a self-assembled monolayer. The self-assembling was carried out in the argon protected environment.

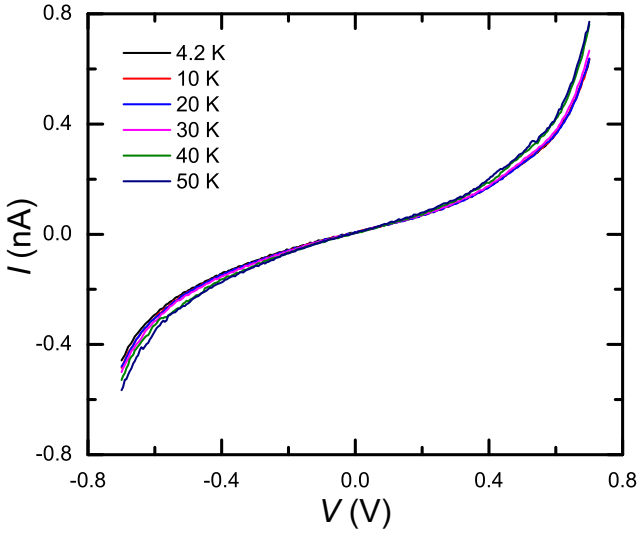

FIG. S2: **Temperature dependence of *I-V* characteristics of a BDF single-molecule device.** The *I-V* curves were measured at different temperatures (4.2 K, 10 K, 20 K, 30 K, 40 K, and 50 K). No obvious temperature dependence was observed, suggesting the transport mechanism is tunneling.

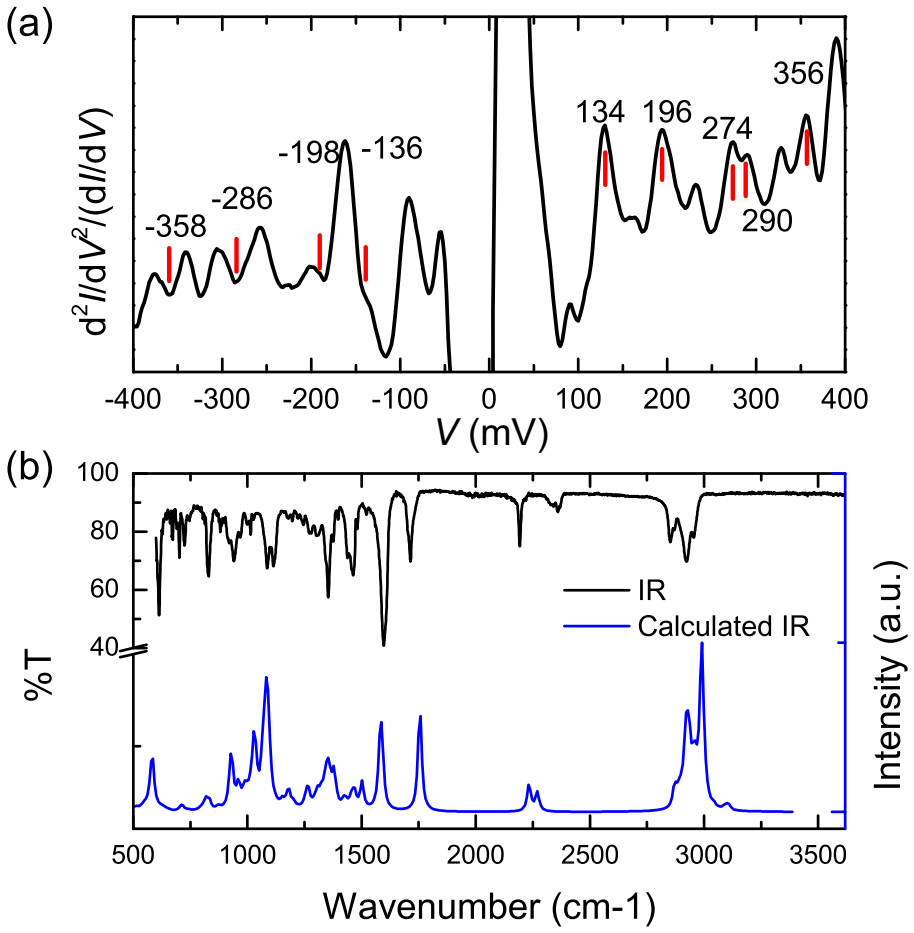

FIG. S3: **Assignment of peaks of IET spectra to specific vibration modes.** **(a)** IET spectra measured at both negative and positive bias voltages. The characteristic signals in the spectrum are highlighted by the red line segments. **(b)** Experimental IR spectrum (black) and DFT calculated IR spectrum (blue) of BDF molecules.

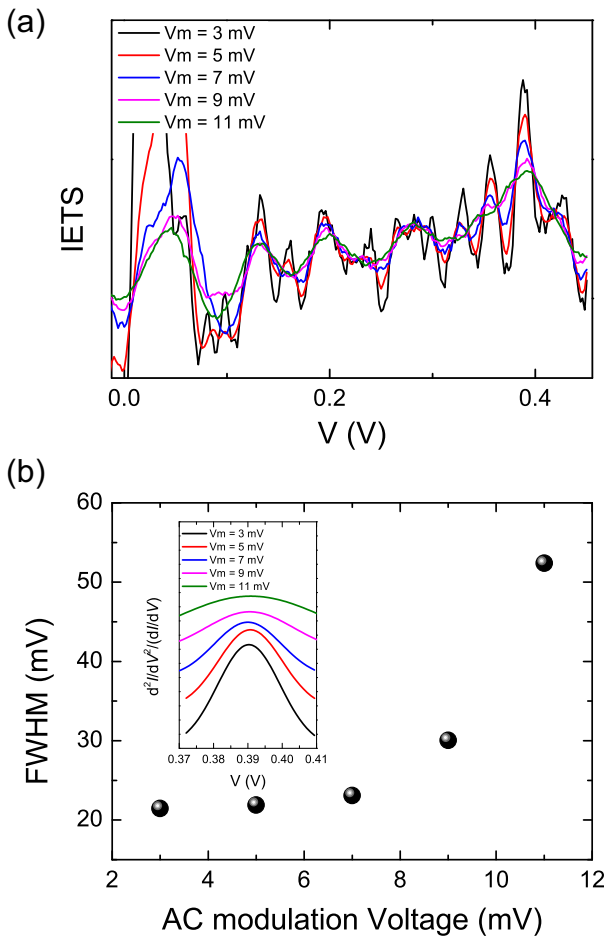

FIG. S4: **The broadening of the ν(C-H) peak (~390 mV, C-H stretch of the phenyl ring) in IET spectra due to the AC modulation.** **(a)** The IET spectra measured at different AC modulation voltages. **(b)** The full width half maximum (FWHM) of the ν(C-H) peak as a function of the AC modulation voltage. Inset: smoothed spectra for the ν(C-H) mode at different AC modulation voltages. The total peak linewidth W consists of three parts: the intrinsic linewidth $W_{intrinsic}$, the thermal broadening $W_{thermal}$ and the modulation broadening $W_{modulation}$. Quantitatively, the total linewidth W can be described by the formula: $W = \sqrt{W_{intrinsic}^2 + W_{thermal}^2 + W_{modulation}^2}$, where $W_{thermal} = 5.4k_BT/e$, and $W_{modulation} = 1.7V_{RMS}$.

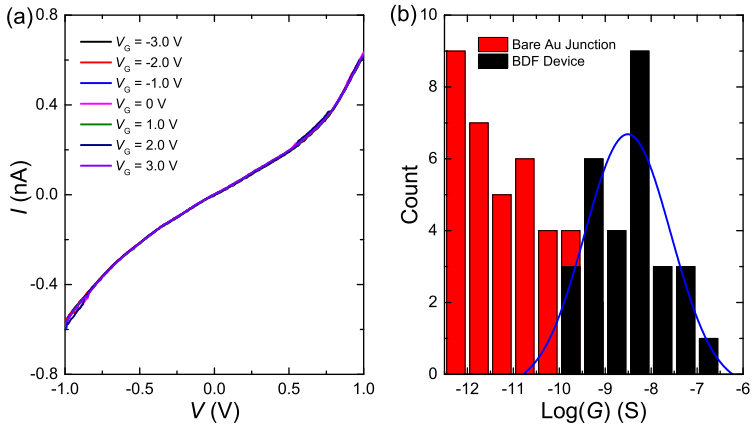

FIG. S5: **The transport properties through gold-vacuum-gold junctions.** **(a)**The *I*-*V* curves of one pristine gold-vacuum-gold junction (without BDF molecules) measured at different gate voltages. **(b)** Comparison of the conductance between gold-vacuum-gold junction and gold-BDF-gold molecular junctions. The controlled experiments were performed under the same condition (4.2 K) as BDF single-molecule devices. In contrast to BDF single molecule devices, the *I*-*V* curves for the control experiments did not show any dependence on the gate modulation.

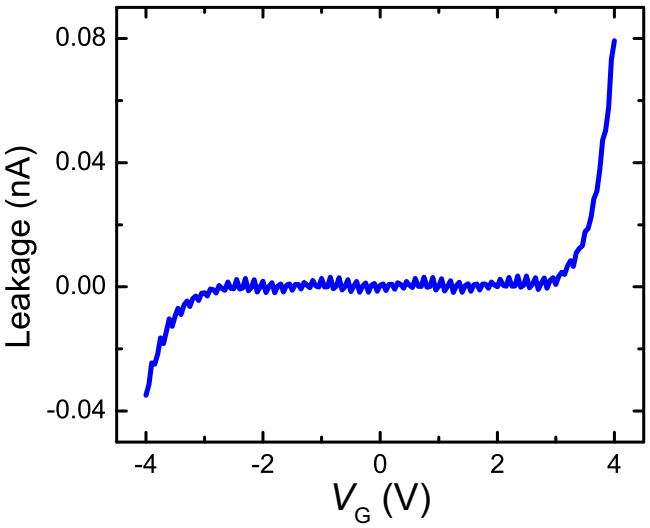

FIG. S6: **Gate leakage in BDF single-molecule transistors.** The leakage current between the source and gate electrodes is below 1 pA in the bias range of 2 V, three orders of magnitude lower than the tunneling current between the source and drain electrodes.

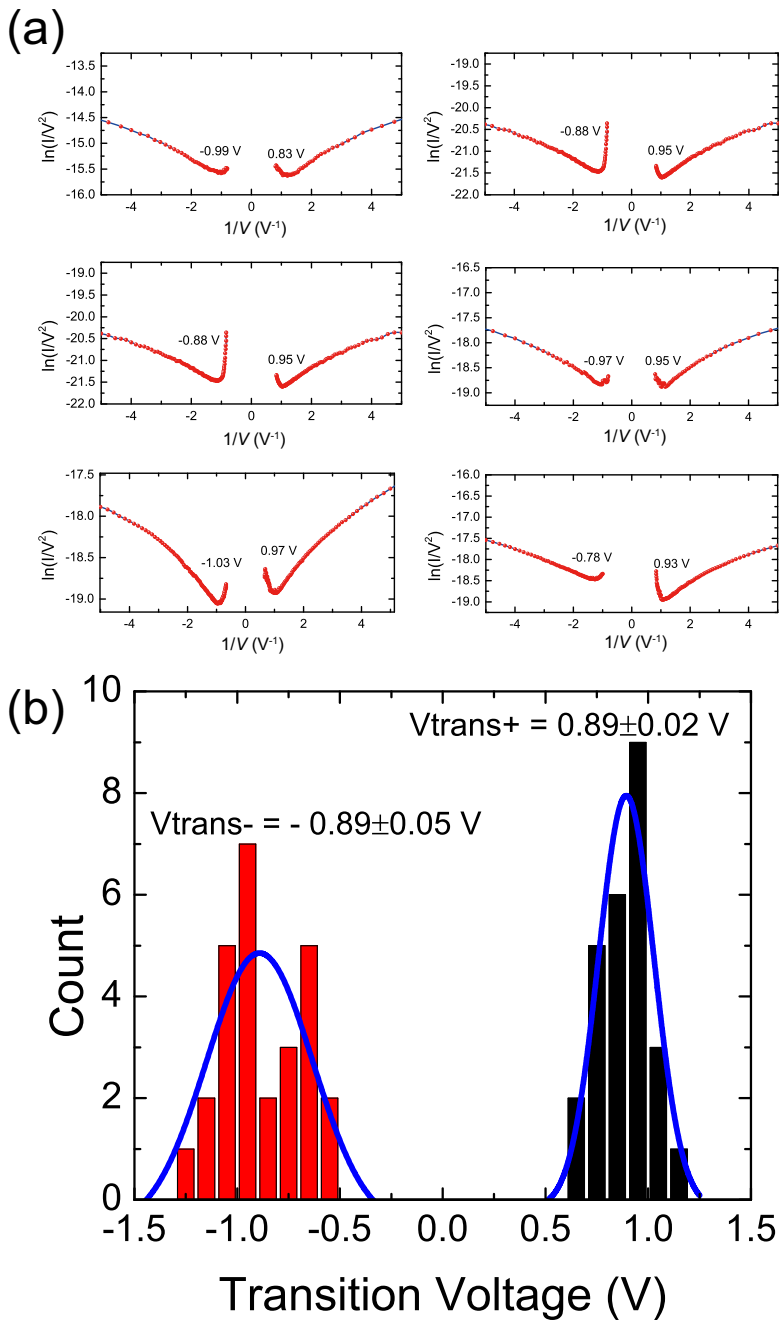

FIG. S7: **Histogram of the transition voltages at two bias polarities for BDF single-molecule devices. a,** F-N plot of *I-V* curves for several devices selected randomly at both polarities. **b,** The transition voltages obtained from Gaussian fitting (blue line) are -0.89±0.05 V and 0.89±0.02 V for the negative and positive bias voltages, respectively. 29 devices were measured.