Single Electron Tunneling – Examples

“It has long been an axiom of mine that the little things are infinitely the most important”
Sir Arthur Conan Doyle

Danny Porath 2003

(Schönenberger et. al.)
Books and Internet Sites

“Electronic Transport in Mesoscopic Systems”, S. Datta

“Single Charge Tunneling”, H. Grabert & M.H. Devoret

“Introduction to Mesoscopic Physics”, Y. Imry

http://www.iue.tuwien.ac.at/publications/PhD%20Theses/wasshuber/
http://vortex.tn.tudelft.nl/grkouwen/reviewpub.html
http://www.ee.princeton.edu/~chouweb/newproject/research/QDT/SingElecTunnel.html
http://qt.tn.tudelft.nl/publi/papers.html
www.unibas.ch/phys-meso
Homework 8

1. Read the paper:
   “Variation of the Coulomb staircase in a two-junction system by fractional electron charge”,

2. Read the paper:
   “Fabry-Perot interference in a nanotube waveguide”,

3. Read the paper:
   “Kondo physics in carbon nanotubes”,
Outline SET Examples:

1. The $C_{60}$ work – D.P. et. al.
   a. Background
   b. Sample preparation and imaging
   c. SET Theory
   d. I-V Spectroscopy results and fits
   e. Summary (of this part)

2. Advanced material from C. Schönenberger
   a. Introduction
   b. Law transparency – SET
   c. High transparency – Fabry Perot & UCF
   d. Intermediate transparency – Co-tunneling & Kondo
   e. Conclusions (from this part)
Single Electron Tunneling and Level Spectroscopy of Single C$_{60}$ Molecules

Danny Porath, Muin Tarabia, Yair Levi and Oded Millo
The General Experimental Scheme
$C_{60}$ Fullerenes

Structure: (icosahedral symmetry)

- 60 carbon atoms. (20 hexagons, 12 pentagons)

$\sim 1$ m $\rightarrow$ 1,000,000,000 $\rightarrow$ $\sim 10$ Å
Background

Before……

- Significant effort was devoted to studies of the interplay between Single Electron Tunneling (SET) effects and quantum size effects in isolated nano-particles.

- The interplay was studied for:
  \[ \Delta E_l \ll E_c \]
  (Tinkham, Van-Kempen, Sivan, Molenkamp)

- C_{60} spectroscopy was done mainly by optical means, limited by selection rules.
**Background**

**What was new here?**

- Extremely small QD (~8 Å)
  - Quantum Size effects even at RT.
- Interplay between SET and discrete levels in two regimes:
  - $E_c < \Delta E_l$
  - $E_c > \Delta E_l$
- Tunneling spectroscopy of isolated $C_{60}$
  - Electronic level splitting.
Sample Preparation

Steps:

- Rinse $C_{60}$ powder in Toluene
- Evaporate dried powder on a gold substrate covered by carbon and (later) a PMMA layer
STM Images of $C_{60}$ Fullerenes
STM Images of C_{60} Fullerenes 3-D
SET effects can be observed If:
1) $E_C = \frac{e^2}{2C} > k_B T$ Charging Energy > Thermal Energy
2) $R_T > R_Q = \frac{h}{e^2}$ Tunneling Resistance > Quantum Resistance

Coulomb Blockade (CB):
$I_T = 0$ up to $|V_t| \leq \frac{e}{2C}$; $V_t$ depends on “fractional charge” $Q_0$

Coulomb Staircase (CS):
Sequence of steps in I-V. Each step - adding an electron to the island
Charge Quantization Leads to SET Effects

Charge quantization leads to SET effects:

- $V_t$ depends on “fractional charge” $Q_0$.
- $Q_0$ represents any offset potentials.
- $Q = ne - Q_0$ ; $|Q_0| \leq e/2$

Allowed states with $n$ access electrons indicated on the graphs
Origin of the Fractional Charge \( Q_0 \)

Difference in contact potentials across the junctions:

\[
Q_0 = (C_1 \Delta \phi_1 - C_2 \Delta \phi_2)/e
\]

Note: We are interested only in \( Q_0 \) mod \( e \).

\( Q_0 \) can be varied by controlling \( C_1 \) through tip-sample separation. This is done by changing the STM current and bias settings.
Interplay Between SET Effects and Discrete Energy Levels

$\Delta E_L \ll E_C$: (Level Spacing $\ll$ Charging Energy)

Additional sub-steps on top of a CS step

(Amman et al.)
Interplay Between SET Effects and Discrete Energy Levels

$\Delta E_L \sim E_C$: (Level Spacing $\sim$ Charging Energy)

- CS steps are no more equidistantly spaced.
- zero bias gap is not suppressed for $Q_0 = e/2$
  when the CB is suppressed.
- Pronounced asymmetry of I–V curves due to different levels on each side.
**I-V Measurements**

- **I-V measurements** are done while disconnecting the feed-back loop.
- **Note:**
  1. Non-vanishing gap.
  2. Pronounced asymmetry and different steps.
  3. Negative differential resistance (NDR).
$I-V$ Measurements

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure.png}
\end{figure}
**HOMO+1 (?)**

- **Diagram 1:**
  - Graph showing the differential conductance ($dI/dV$) vs. tip voltage ($V$).
  - Peaks indicating transitions through different energy levels.

- **Diagram 2:**
  - Energy level diagram with states labeled HOMO, LUMO, and LUMO+1.
  - Fermi level ($E_F$) and transitions through LUMO and LUMO+1.

- **Diagram 3:**
  - Tunneling process through LUMO+1 indicated with arrows.

- **Diagram 4:**
  - Tunneling process through LUMO with note: $n=2$ on the dot.
Different electric fields (carbon field, STM tip) effect the molecular orbitals differently (Stark effect).
Molecule Orientation (Guntherodt et. al.)

Tunneling channel (depending on the molecule orientation) effects the level splitting.
Jahn - Teller effect (Bendale et. al.)

- A splitting or a shift of the energy levels due to structural distortion.
- The $C_{60}^+$ $h_u$ reduces by $\sim 0.6$ eV
Theoretical Fits (Orthodox theory-Averin & Likharev)

Schematic of the circuit:

The tunneling rate on the dot:

\[ \Gamma_i^{\pm}(n) = \frac{2\pi}{\hbar} \int \left| T_i(E) \right|^2 D_i(E - E_i) f(E - E_i) D_d(E - E_d) [1 - f(E - E_d)] dE \]

- \( \Gamma_i^{\pm}(n) \) - Tunneling rate to/from the dot from/to the electrode.
- \( f \) - Fermi function.
- \( D_i(E), E_i \) - DOS/Fermi energy at electrode i.
- \( D_d(E), E_d \) - DOS/Fermi energy at the dot.
- \( T_i(E) \) - Tunneling matrix element.
Theoretical Fits (Orthodox theory-Averin & Likharev)

We choose the density of states $D_d(E)$:

\[ \sum \Gamma (n) P(n) \]

The tunneling current is:

\[ I(V) = e \sum P(n) \left[ \Gamma_2^+(n) - \Gamma_2^-(n) \right] = e \sum P(n) \left[ \Gamma_1^-(n) - \Gamma_1^+(n) \right] \]

\[ \text{P}(n) - \text{The probability to find n electrons on the dot} \]
Energy Level Calculations
Experimental Results – Asymmetrical Curves

$$C_1 \sim C_2 \sim 10^{-19}, \ E_{\text{HOMO}} - E_{\text{LUMO}} \sim 0.7 \text{ eV}, \ \Delta E_L \sim 0.05 \text{ eV}$$
Source of Asymmetry

$C_1 < C_2 \implies V_1 > V_2 \implies$ Onset of tunneling at junction 1 (for the level configuration at hand)

Positive bias

Negative bias
Experimental Results – Symmetrical Curves

\[ C_1 \sim C_2 \sim 10^{-19}, \ E_{\text{HOMO}} - E_{\text{LUMO}} \sim 0.7 \text{ eV}, \ \Delta E_L \sim 0.05 \text{ eV} \]
Symmetrical Barriers \((C_1 \sim C_2)\)

- **Positive bias**
- **Negative bias**

- Tunneling through the set of levels closer to \(E_F\) - in our case the 3 LUMO levels.
Checking The Model - Experimentally

\[ C_1 < C_2 \]
\[ I_s = 1.6 \text{ nA} \]
\[ V_s = 0.75 \text{ V} \]

\[ C_1 > C_2 \]
\[ I_s = 6 \text{ nA} \]
\[ V_s = 0.75 \text{ V} \]
The Triple Barrier Tunnel Junction (TBTJ)

Note:
- Non Vanishing gap.
- Negative differential resistance (NDR).
- Non-ohmic curves everywhere on the sample.
Summary - $C_{60}$ STM Spectroscopy

A $C_{60}$ molecule is an ideal “quantum dot” for studies of single electron tunneling and quantum size effects.

The interplay between SET effects and molecular levels is clearly observed at 4.2 K as well as at RT for the double barrier tunnel junction configuration.

The degeneracy of the HOMO, LUMO and LUMO+1 levels was fully resolved, and the degree of splitting is consistent with the J-T effect.
Quantum Dot Physics

C. Schönenberger
Quantum dots

What are Quantum Dots?

Quantum dots are nanometer ($10^{-9}$ meter) scale particles that are neither small molecules nor bulk solids. Their composition and small size (a few hundred to a few thousand atoms) result in extraordinary optical properties that can be readily customized by changing the size of the dot, or composition of the dot material, or both. When placed in a different environment, the particles can exhibit a phenomenon known as quantum confinement. By matching with this phenomenon can be excited by a light wave

Quantum dots can be made from a wide variety of materials, such as semiconductors, metal, and organic molecules, and can be linked to other materials such as gold nanoparticles. There are many applications of interest, such as sensing and fluorescence, and many are for making light emitting devices (LEDs), lasers, or organic quantum dots that are found in a variety of applications. Quantum dots can be fabricated in laboratories using a variety of methods, including quantum dots emitting light of different wavelengths. By using only a small number of quantum dots emitting light of different wavelengths, LED can be made that are found in a variety of applications. Quantum dots with colors that span the spectrum, from ultraviolet to infrared.
Quantum dots are nanometer-scale "boxes" for selectively holding or releasing electrons. Over the past 10 years they have been transformed from laboratory curiosities to the building blocks for a future computer industry.
quantum dot?
Different energies (what is 0d?)

- Standing waves (particle in a box)
- Discrete spectrum
- Level-spacing $\delta E$

In addition:
- $eV$ and $kT$

Example: 0d with respect to quantum-size effects, provided: $kT$, $eV$, and $\Gamma < \delta E$
Contacts matter...!

1. **low transparency** → single-electron tunneling determined by **single-electron charging** effects, e.g. **Coulomb blockade** (0d limit)

2. **intermediate transparency** → charging effects but **co-tunneling**, e.g. **Kondo effect**

3. **high transparency** → quantum interference of **non-interacting electrons**, e.g. **Fabry-Perot resonances** and **UCF** (disordered limit)
Contacts matter...!

1. low transparency $\rightarrow$ single-electron tunneling determined by single-electron charging effects, e.g. Coulomb blockade (0d limit)
Single-electron tunneling

Here: black = low differential conductance

Coulomb blockade
tutorial on dI/dV plots

\[ \Delta E_{\text{add}} \text{ addition energy, i.e. sum of:} \]

- single-electron charging energy \( U_C \)
- level-spacing \( \delta E \)

\[ \Delta E_{\text{add}} = U_C + \delta E \]

Change \( V_{sd} \)

\[ eV_{sd} \]

Change \( V_g \)

\[ \Gamma_S \quad \Gamma_D \]

\[ \mu_S \quad \mu_D \]
filling of states according to $S = \frac{1}{2} \rightarrow 0 \rightarrow \frac{1}{2}$ ...

**odd number of electrons:**

$$\Delta E_{\text{add}} = U_C$$

**even number of electrons:**

$$\Delta E_{\text{add}} = U_C + \delta E$$
single-electron-tunneling

if tunneling probability $p$ of each junction is „small“:

„uncorrelated“ sequential tunneling dominates. Current $I \propto p$

current determined by accessible levels in dot
i.e. by level spacing and Coulomb charging energy
Contacts matter...!

1. low transparency $\rightarrow$ single-electron tunneling determined by single-electron charging effects, e.g. Coulomb blockade (0d limit)

2. intermediate transparency $\rightarrow$ charging effects but co-tunneling, e.g. Kondo effect

3. high transparency $\rightarrow$ quantum interference of non-interacting electrons, e.g. Fabry-Perot resonances and UCF (disordered limit)
T~1 limit (interaction-free wire)

Fabry–Perot interference in a nanotube electron waveguide

Wenjie Liang*, Marc Bockrath†, Dolores Bozovic‡, Jason H. Hafner*, M. Tinkham‡ & Hongkun Park*

* Department of Chemistry and Chemical Biology and ‡ Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA
† These authors contributed equally to this work

5 mV -5 mV

bias voltage

-2 ... +2 V

gate voltage

T~1 limit (wire)

\[ T(E) = \frac{T_C}{1 + R_C^2 + 2R_C \cos(\Theta + 2\delta_R)} \]

\[ \Theta = L(k^+ - k^-) \quad \text{round-trip phase depends on } E \]

\[ \frac{\partial I}{\partial V} = \frac{e}{2} \{T(eV/2) + T(-eV/2)\} \]

Fabry-Perot (interference)

gate voltage -2 ... +2 V

5 mV
-5 mV

their data

simple model
Contacts matter...!

1. **low transparency** → single-electron tunneling determined by **single-electron charging** effects, e.g. Coulomb blockade (0d limit)

2. **intermediate transparency** → charging effects but **co-tunneling**, e.g. Kondo effect

3. **high transparency** → quantum interference of **non-interacting electrons**, e.g. Fabry-Perot resonances and UCF (disordered limit)
if tunneling probability $p$ of each junction is "small":

"uncorrelated" sequential tunneling dominates. Current $I \propto p$

if tunneling probability $p$ of each junction is "large":

coherent 2nd (and higher) order processes add substantially $\propto p^2$

we call this "co-tunneling"
Remainder Coulomb blockade

black regions =
very low conductance $G$

$G$ is suppressed due to
Coulomb blockade (CB)

$I$ jump back to low transparent tunneling contacts for reference
Elastic co-tunneling

\[ \Delta E_{\text{add}} = U_C + \delta E \]

\[ \mu_S \rightarrow \mu_D \]

Inelastic co-tunneling

\[ \Delta E_{\text{add}} = \mu_D - \delta E \]

\[ \mu_S \rightarrow \mu_D \]

\[ eV_{sd} = \delta E \]
When the number of electrons on the quantum dot is odd, spin-flip processes (which screen the spin on the dot) lead to the formation of a narrow resonance in the density-of-states at the Fermi energy of the leads.

This is called the Kondo effect

Kondo effect

**Resistance** $R(T)$ of a piece of metal

- "ideal" metal, e.g. Au wire
- Superconductor, e.g. Pb
- Magnetic impurities in "ideal" metal, e.g. (ppm)Fe:Au → Kondo system

**Conductance** $G(T)$ through a single magnetic impurity (e.g. an spin $\frac{1}{2}$ quantum dot)

- Unitary limit $2e^2/h$
- $T_K$ at ~0.5 K
- $-\log(T/T_K)$ at ~10 K
S=1/2 Kondo in Q-dots

from L. Kouwenhoven & L. Glazman, Physics World, June 2001
at 50 mK

Mark Buitelaar and Thomas Nussbaumer
why interesting?

because it is complicated 😊

it’s many-body physics (all orders are relevant)

more precisely: it’s many-electron physics

in Condensed Matter Physics:

- Superconductivity
- Superfluidity
- Luttinger liquid (non-Fermi liquids)
- Kondo physics
Kondo effect and superconductivity are many-electron effects

• can Kondo and superconductivity coexist or do they exclude each other?
Kondo effect is the screening of the spin-degree of the dot spin by exchange with electrons from Fermi-reservoirs (the leads).

normal case

superconducting case

1. a gap opens in the leads
2. Cooper pairs have S=0

Hence: Kondo effect suppressed, but ....
A cross-over expected at $k_b T_K \sim \Delta$
Conclusions

nanotubes may be one part of the toolbox of „nanotechnology“

nanotubes can serve as a model system to study exciting physics

- Kondo physics (co-tunneling)
- Interplay between Kondo physics & superconductivity

www.unibas.ch/phys-meso

www.nccr-nano.org

NCCR on Nanoscience