Non-linearities in quantum circuits

Serge Florens [Néel Institute - CNRS/UGA Grenoble]

“A gentle journey into not-so-gentle quantum many-body problems”
Organization of the lectures

Superconducting nanocircuits

▸ Lecture 1: i) General circuitry; ii) From Josephson junctions chains to spin-boson model

▸ Lecture 2: i) Scattering theory with dynamical emitter; ii) Frequency conversion beyond RWA; iii) Renormalization from many-body entanglement; iv) Many-body inelastic scattering theory

Metallic nanocircuits

▸ Lecture 3: i) Anderson and Kondo models; ii) DC Transport; iii) Non Fermi liquid extensions

▸ Lecture 4 (closing the loop): i) Boson-fermion connection in first quantization (using quantum optics concepts); ii) Insights into the Kondo wave-function; iii) Emergent Majorana fermions
Various quantum dot systems
Rules of the game

What kind of experimental system does one need?

1. Good tunability
2. Reasonable energy scales

Reminder: Helium dilution fridges work for $T \gtrsim 20\text{mK}$

$(1\text{K} \simeq 0.1 \text{meV} \simeq 20 \text{GHz})$
Semiconducting quantum dots

- **System**: 2D electron gas (GaAs, InAs, S, Ge...)
- **++ great tunability and scalability by electric gates**
- **−− small charging (Coulomb) energy**:
  - $U = 40K$ for spin qbit experiments
  - $U < 10K$ for Kondo experiments
  
  Ultra-small Kondo temperature $\Rightarrow T_K < 300mK$

![Experimental systems](image)

: Non-linearities in quantum circuits
Experimental systems

Carbon nanotube quantum dots

- System: 1D carbon molecule with gates on top
- ++ tunable & hybrid: metal, ferro or superconducting leads
- + intermediate charging energy $U = 10K - 100K$
- ± orbital degeneracy: SU(4) vs SU(2) physics

Ferro [ENS-Paris]

SQUID [Grenoble]
Molecular devices

- System: nanometer size molecule inbetween metal contacts
- Limited tunability (no local gates, but some surprises...)
- ++ more tunability from chemistry
- −− lack of reproducibility
- ++ very large energy scales $U = 1000K - 10000K$
  $\Rightarrow$ good playground for Kondo physics!
Molecular electronics HOWTO : electromigration

Basic idea:

- E-beam lithography cannot reach single molecule size
- Solution: open a metal junction via electromigration
- Back gate: molecular transistor
Molecular electronics HOWTO: break junctions

Basic idea:

- Solution: open mechanically a metal junction
- No back gate, but strain changes tunnel couplings
Overview of regular (screened) Kondo effect
Kondo chronology

Textbook stuff in solid state physics:
- 1934: observation of resistance anomalies in metals
- 1964: Kondo proposes his model
- 1975: Wilson solves the “Kondo problem”

The Kondo revival:
- 1988: Glazman-Raikh & Ng-Lee predict Kondo anomalies in quantum dots
- 1998: First observation by Goldhaber-Gordon & Kouwenhoven
- Now: it is routinely observed in all kinds of nanostructures!

Some good references to dig deeper:
- Coleman: “Heavy electron systems” cond-mat/0206003
- Glazman&Pustilnik: “Les Houches 2004” cond-mat/0501007
The essence of the Kondo problem

**Question**: what becomes a magnetic atom in a metal?

**Experimental fact**: magnetic impurities not always show a moment

**Simplified atom**: \( H_U = \epsilon_d (n_{d\uparrow} + n_{d\downarrow}) + Un_{d\uparrow}n_{d\downarrow} \)

\( \Rightarrow \) Moment stabilized by \( U \), high energy excitations

**Resonant level**: \( H_t = \sum_{k\sigma} [\epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + td_{\sigma}^\dagger c_{k\sigma} + h.c] \)

\( \Rightarrow \) No moment, resonant level at low energy

**Anderson model**: \( H = H_U + H_t \)

\( \Rightarrow \) Competition localized/delocalized

Captures the physics between the two above limits
On the board: Atomic limit $t = 0$
Local moment formation: $t \neq 0 \ (\epsilon_d = -U/2 \ here)$

Mean field theory \[\text{[Anderson 1950]}\] :
Assumes a frozen moment $\mu = \langle n_{d\uparrow} - n_{d\downarrow} \rangle$

Results:
- Local moment $\mu \neq 0$ for $U > \pi \Gamma = \pi t^2 \rho_0$
  $\Rightarrow$ need relatively closed quantum dot
- Broadened high energy features
  (Hubbard bands $\Leftrightarrow$ Coulomb blockade edges)
Coulomb blockade: weakly open quantum dot

**Conductance vs. gate:**

- Charging energy dominates: \( H = \frac{U}{2} (Q_{\text{dot}} - \alpha e V_g / U)^2 \)
- Frozen charge fluctuations: transport suppressed
- Conductance has peaks at charge degeneracy
Coulomb blockade: weakly open quantum dot

Conductance vs. gate and bias:

- Coulomb diamonds with low conductance
- Totally generic observation!
- Single charge transistor
Coulomb blockade suppression: Kondo anomaly

Moderately open quantum dot:

Zero-bias transport is restored for odd charge
Physical origin of Kondo effect

- General idea: lifting of a degeneracy by a Fermi sea through exchange interaction
- Odd charge quantum dots: resonant spin flip processes

- Resonant binding of electrons near Fermi level
- Open channel for transport

Theory: Glazman & Raikh, Ng & Lee (1988)

Experiment: Goldhaber-Gordon, Kouwenhoven (1998)
On the board: perturbation at order $U^2$
Fermi liquid description [Langreth, Nozières, Yosida,...]

Small $U$: perturbation theory in $U$ well-behaved

⇒ Self-energy: $\Sigma_d(\omega) = (1 - Z^{-1})\omega + iA\omega^2$ for $\omega \to 0$

⇒ Effective width: $T_K = Z\Gamma$ Renormalized quasiparticles

Conductance from linear-response: Landauer-like

$$G(T) = \frac{2e^2}{h} \frac{4t_L^2 t_R^2}{(t_L^2 + t_R^2)^2} \int d\omega \ n'_F(\omega) \pi \Gamma \text{Im}[G(\omega)]$$

Friedel sum rule: $\text{Im}[G(0)] = \frac{1}{\Gamma}$ at $T = 0$ and $\epsilon_d = -U/2$

⇒ $G(T = 0) = \frac{2e^2}{h}$ for symmetric barriers

Key question: does that remain true for $U \gg \Gamma$?
On the board: Schrieffer-Wolff transformation
On the board: Schrieffer-Wolff transformation

Schrieffer-Wolff transformation: keep only spin states at large $U \
\Rightarrow$ Antiferromagnetic coupling $J_K = 8\frac{t^2}{U}$ to the Fermi sea

Kondo model:

$$H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + J \vec{S} \cdot \sum_{kk'\sigma\sigma'} c_{k\sigma}^\dagger \frac{\vec{t}_{\sigma\sigma'}}{2} c_{k'\sigma'}$$

$$H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + J \vec{S} \cdot \sum_{\sigma\sigma'} c_{\sigma}^\dagger (x = 0) \frac{\vec{t}_{\sigma\sigma'}}{2} c_{\sigma'}(x = 0)$$
Kondo logarithms \cite{Abrikosov, Suhl, Nagaoka, Kondo, Anderson...}

Perturbation in $j = J/D$ : logarithmic divergent at low $T$

$$\chi_{spin}(T) = \frac{3}{4T} \left[ 1 - j - j^2 \log \frac{D}{T} + \ldots \right] \simeq \frac{3}{4T} \left[ 1 - j_R(T) + \ldots \right]$$

$$G(T) = \frac{2e^2}{h} \frac{3\pi^2}{16} \left[ j^2 + j^3 \log \frac{D}{T} + \ldots \right] \simeq \frac{2e^2}{h} \frac{3\pi^2}{16} \left[ j_R^2(T) + \ldots \right]$$

- Kondo temperature:
  $$T_K = De^{-D/J} = De^{-\pi U/8\Gamma}$$

- Renormalized coupling:
  $$j_R(T) = \frac{1}{D/j + \log(T/D)} = \frac{1}{\log(T/T_K)}$$
  grows

- Hints that local moment does not survive at $T \ll T_K$
Solution of the Kondo problem : an NRG primer

Algorithm : [Wilson (1975)]

▶ Logarithmic discretization of the Fermi sea : \([\Lambda^{-k-1}, \Lambda^k]\)
▶ Mapping of a chain with exponential vanishing couplings
▶ Iterative solution : diagonalize + truncate Hilbert space

Remarks :
▶ Resolves low energy features (Kondo peak)
▶ Stability requires scale separation : \(\Lambda > 2\)
▶ Discretization errors \(\sim e^{-\pi^2 / \log(\Lambda)} \rightarrow \text{small!}\)
Illustration of the universal crossover

Curie constant and entropy from NRG calculations:

- Three large values of $U/\Gamma$

Perfect data collapse:

- All observables are functions of $T/T_K$ only

$\Rightarrow$ universality!
Back to the experiments

- Conductance is large at $T \lesssim T_K$: Kondo prevails
- Coulomb blockade is recovered at $T \gg T_K$

General Friedel sum rule at $T=0$:

$$G = \frac{2e^2}{h} \frac{4 t_L^2 t_R^2}{(t_L^2 + t_R^2)^2} \sin^2 \left(\frac{\pi}{2} Q_{\text{dot}}\right)$$
Testing the theory with experiments: $G(B)$ and $G(T)$

**C$_{60}$ molecular device** [Scott et al. PRB 2013] (Rice)

Successful fit of $G(T)$ and $G(B)$ with consistent $T_K \simeq 10K$
Non-Fermi liquid extensions
Spin $S = 1$ : screening versus underscreening

Two screening channels: full quenching $\implies$ regular Kondo effect

One screening channel: residual spin $\implies$ anomalous Kondo effect
Non-Fermi liquid extensions

Under screening: Nozières-Blandin argument

Spin $S = 1$ and single screening channel:

- Effective spin $S_{\text{eff}} = 1/2$ (residual entropy)
- Effective Kondo coupling $J_{\text{eff}} \propto -t^2/J$ : ferromagnetic!
  $$\implies \text{temperature dependence: } J_{\text{eff}}(T) \propto \frac{1}{-J_{\text{eff}} + \log(T/T_0)}$$
  $$\implies J_{\text{eff}}(T) \text{ vanishes at low } T$$

Transport: logarithmic approach to unitarity

$$G(T) = \frac{2e^2}{h} \left[ 1 - \frac{c}{\log^2(T/T_K)} \right]$$

: Non-linearities in quantum circuits
First experimental evidence of underscreening

$C_{60}$ device: [Roch et al., Nature (2008)] (Grenoble)

- Surprising gate effect on the singlet-triplet gap
- All transport signatures now well-understood
A second experiment: break junction device

Cobalt in a “cage”: [Parks et al. cond-mat (2010)] (Cornell)

Underscreened Kondo anomaly: clearly logarithmic below $T_K$
Two-channel Kondo effect

Spin $S = 1/2$ but two independent Fermi seas ($m=1,2$):

$$H = H_1 + H_2 + \sum_{kk'} \sum_{\sigma\sigma'} \sum_m J_m c_{k\sigma m}^\dagger \frac{\vec{t}_{\sigma\sigma'}}{2} c_{k'\sigma' m} \cdot \vec{S}$$

Strong coupling argument: at $J_1 = J_2$ large

- Effective spin $S_{\text{eff}} = 1/2$ (residual entropy)
- Effective Kondo coupling $J_{\text{eff}} \propto +t^2/J$: antiferromagnetic!

$\implies$ strong coupling fixed point is unstable!
Quick status of 2CK : theory and experiment

Some theoretical facts : (NRG, CFT, Bosonization…)

- $S(T=0) = \log(\sqrt{2}) = \log(2)/2$ : emergent Majorana !
- $\chi(T) \propto \log(T_K/T)$
- $G(T)/G_0 \simeq 1 - a\sqrt{T/T_K}$ : non Fermi liquid

Several experimental studies : (discussion beyond this course)

[Ifthikar et al. Nature 2015] (LPN)
Extra slides
Non-Fermi liquid extensions

Nice illustration with “real” and “fake” spin 1/2

Even charge CNT quantum dot: [Nygard et al., Nature (2000)]

- Magnetic-field induced degeneracy by crossing of singlet and lowest triplet

\[ E_{11,0} \] \[ E_{11,-1} \] \[ E_{10,0} \]

\[ |0,0\rangle \] \[ |1,-1\rangle \]

\[ |0,0\rangle = |\downarrow\rangle \] \[ |1,1\rangle = |\uparrow\rangle \]
Another illustration with “fake” spin

Charge states in double dot setup:

- Pseudospin: $(1,0) \leftrightarrow \uparrow$ and $(0,1) \leftrightarrow \downarrow$
- Kondo process takes place

[Goldhaber-Gordon group (2012)]
How to reach the scaling limit?

Universal (sharp) Kondo resonance requires:
- well-formed local moment i.e. large $U/\Gamma$

Consequence: only in the Kondo regime
- single parameter scaling of physical quantities
  Zero-bias Conductance: $G(T) = G_0 \ast f(T/T_K) + G_1$

Question for experiments: how to quantify the universal regime?
Precise condition to be universal

**Wilson ratio**: \( R = \frac{\chi}{\gamma} \times \frac{\gamma}{\chi|_{U=0}} \)

- **Zero** \( T \) spin susceptibility (screening): \( \chi(T) \propto \frac{1}{T_K} \)
- **Low** \( T \) specific heat (Fermi liquid): \( C(T) = \gamma T \propto \frac{T}{T_K} \)

**Effective width**:

\[
\tilde{\Delta} / \Delta \approx 4T_K / \pi \Delta
\]

**Kondo scale**:

\[
T_K \propto e^{-\pi U / 8\tilde{\Gamma}}
\]

- 3% deviation to universality: \( U \gtrsim 2\pi \tilde{\Gamma} \Rightarrow T_K \lesssim U / 40 \)
- Also constraints on level spacing
Non-Fermi liquid extensions

Testing the theory with experiments: $G(T)$

Semiconducting quantum dots: [van der Wiel et al. Nature (2000)]

Sizable deviations to scaling due to too small $U/T_K$

Molecular quantum dots: [Roch et al. PRL (2009)]

$T_K = 4K$, $U > 600K$ conditions are met!
Testing the theory with experiments: $G(B)$

Magnetotransport data in molecular quantum dot:

Fit of the Zeeman splitting:

- Kondo peak splits at $B_c = k_B T_K/(2g\mu_B) \Rightarrow T_K = 4.8K$ OK!
Testing the theory with experiments: $G(B)$

Attempt at scaling analysis: (InAs nanowire, Mahalu’s group)

- $G(T)$ and $G(B)$ are different universal scaling functions
- The $T_K$ and $B_K$ scales are not consistent!
- The experiment is quantitatively not fully conclusive...
Out of equilibrium scaling

Finite-bias conductance $G(V)$:

- Main physical effect: enhanced scattering from large current density kills the Kondo resonance
- Tough problem for many-body theory
- Reliable experimental data are needed also

**Methods:** [NRG (Anders 2009), QMC (Han 2007), RTRG (Pletyukhov-Schoeller 2012)]

- $T_K$ and $V_K$ consistently predicted by RTRG
- Deviations to scaling (semiconducting dot)
Checking Fermi Liquid Theory

At low energy: \( T \ll T_K \) and \( eV/k_B \ll T_K \)

\[
G(T, V) = G_0 \left[ 1 - c_T \left( \frac{\pi T}{T_K} \right)^2 - c_V \left( \frac{eV}{k_B T_K} \right)^2 \right]
\]

Out of equilibrium Fermi Liquid theory: [Oguri JPS (2004)]

\[
\alpha \equiv \frac{c_V}{\pi^2 c_T} = \frac{3}{4\pi^2} \frac{1 + 5(R - 1)^2}{1 + 2(R - 1)^2}
\]

\[
= \frac{3}{2\pi^2} \simeq 0.16 \text{ [Kondo regime (} R = 2)\text{]} \\
= \frac{3}{4\pi^2} \simeq 0.08 \text{ [uncorrelated (} R = 1)\text{]}
\]

Note: \( \alpha \) depends on \( \Gamma_L/\Gamma_R \) in general
but not any more in the \( R = 2 \) limit (universality again!)
Hunt for $\alpha$ : 2DEG quantum dot data

Recent experiment : [Grobis et al. PRL (2008)]

Fermi liquid coefficients :

- Nice systematics !
- $0.1 < \alpha < 0.15$ : signature of intermediate correlations ?!
Hunt for $\alpha$ : molecular quantum dot data

Grenoble experiment : [Roch et al. PRL (2009)]

- Here $T_K = 4K \implies$ less noise!
- For this only sample: we extract $\alpha \simeq 0.15$, OK!
- See recent study by Mahalu’s group
Non-Fermi liquid extensions

Illustration: whole crossover in diluted alloy

Resistivity of Kondo compound $\text{Mo}_x\text{Nb}_{1-x}$:

Scaling on 4 orders of magnitude!!
Disgregation: Kondo anomalies in \( \text{Ag}_{1-x}\text{Fe}_x \) and \( \text{Au}_{1-x}\text{Fe}_x \)

Resistance of wires: \[ \text{[Costi et al. PRL 2009]} \]

- LDA predicts spin \( S = \frac{3}{2} \) for Fe and 3 orbitals involved in screening process

Data compatible with full screening (no underscreening!)

Theory cannot distinguish the spin value \( S \)
Disgression: Kondo anomalies in wires

Inelastic contribution to the resistivity:

- This allows to discriminate the spin value:
  \[ S = \frac{3}{2} \text{ and } 3 \text{ orbital involved!} \]

- This systems is thus not underscreened...
Non-Fermi liquid extensions

Identifying the spin states: gate voltage scan at $B = 3\, \text{T}$

- Zeeman effect agrees with spin 0 or spin 1 ground state
- Gate-induced magnetic splitting (tunable Hund’s rule!)

: Non-linearities in quantum circuits
Non-Fermi liquid extensions

Origin for the gating effect: phenomenology

Role of the leads: energy gain by charge fluctuations

Hopping from level 1 or 2 \[ \rightarrow \delta E_{1,2} = -\frac{t_{1,2}^2}{E_{add}} \]

Hypothesis: hopping asymmetry \[ t_1 \gg t_2 \] explains data

Crucial observation: single screening channel!!
Non-Fermi liquid extensions

Second evidence for single screening channel

Magnetic field effect:

- No Kondo anomaly at the singlet-triplet degeneracy point!

Kondo coupling:

\[ J_K \propto \frac{t_1 t_2}{E_c} \text{ small as } t_1 \gg t_2 \]
Modelization the gating effect

Generalized two-orbital Anderson model:

\[
H = \sum_{i\sigma} E_i n_{i\sigma} + \sum_i U_{i} n_{i\uparrow} n_{i\downarrow} + U_{12} n_1 n_2 - J_H s_1 s_2
\]

\[
+ \sum_{\nu k \sigma} \epsilon_{\nu k} c_{\nu k \sigma}^{\dagger} c_{\nu k \sigma} + \frac{1}{\sqrt{N_k}} \sum_{i \nu k \sigma} (t_{i \nu} c_{\nu k \sigma}^{\dagger} d_{i \sigma} + \text{h.c.})
\]

with \( E_2 - E_1 \simeq J_H / 2 \) (small bare singlet-triplet gap)
and \( t_1 \simeq 2 \times t_2 \) (moderate hopping asymmetry)

Scenario confirmed by NRG: [Florens et al., JPCM (2011)]
Testing the underscreened Kondo scenario

Analysis of the spin $S = 1$ Kondo anomaly: [Roch et al., PRL (2009)]

- Agreement with $S = 1$ NRG clearly better
  ... but tough experiment!!
- $\frac{dG(T)}{dT}$ shows two logarithmic regimes
Non-Fermi liquid extensions

Magnetic field effect: smoking gun!

Comparing $S = 1/2$ and $S = 1$ Kondo anomalies:

- Splitting occurs at lower magnetic fields for $S = 1$

NRG calculations: $S = 1$ Kondo resonance very sensitive to field
Non-Fermi liquid extensions

Analysis of the transition

- Far on the singlet side: spin gap
- Far on the triplet side: underscreened Kondo
- Nature of QCP:
  one free spin $S = 1/2 \oplus$ one screened spin $S = 1/2$
Non-Fermi liquid extensions

Around the quantum critical point

Singlet side:

Triplet side:

- For $T \gg |E_T - E_S|$ (on both sides): broad spin $S = 1/2$ Kondo anomaly
- Sharp antiresonance (Friedel !) on the singlet side at low $T$ $\implies$ second-stage of Kondo screening
Unexpected effect

Stretching the molecule: splits the Kondo resonance!

Interpretation: strain induced magnetic anisotropy?

Putting the $\left| 1, 0 \right>$ state down: kills underscreened Kondo effect
Where are we in parameter space?

Travelling along Anderson’s flow diagram:

- Cornell
- Grenoble

Diagram showing the flow between ferro and anti-ferro phases with parameter axes $J_\perp$ and $J_{//}$.
A third experiment: semiconducting dots

**Obvious way to tune QPT**: just change the charge!

- $S = 1/2$: Fermi liquid, sum rule $\sin^2(\pi Q/2)$
- $S = 1$: non-Fermi liquid, sum rule $\cos^2(\pi Q/2)$

[Pustilnik & Borda PRB 2006; Logan et al. PRB 2009; Kogan et al. PRB 2003]

QPT signaled by conductance jump at $T = 0$
Channel anisotropy kills 2CK

Weak coupling RG: with $J_1 \neq J_2$

$$H = H_1 + H_2 + \sum_{\sigma \sigma'} \sum_m J_m \, c_{\sigma \, m}^\dagger \frac{\vec{T}_{\sigma \sigma'}}{2} c_{\sigma' \, m} \cdot \vec{S}$$

$\implies$ fine tuning needed!

but can one realize such Hamiltonian with quantum dots?
Non-Fermi liquid extensions

Why 2 leads experiments give only 1 channel?

Kondo Hamiltonian: $\alpha = L, R$

$$H = H_L + H_R + \sum_{\sigma\sigma'} \sum_{\alpha,\alpha'} J_{\alpha,\alpha'} c_{\sigma\alpha}^{\dagger} \frac{\vec{\tau}_{\sigma\sigma'}}{2} c_{\sigma'^{\prime}\alpha'} \cdot \vec{S}$$

Matrix of couplings: $J_{\alpha,\alpha'} \propto \frac{t_{\alpha} t'_{\alpha}}{U}$ has only one non-zero eigenvalue

$$\Rightarrow H = H_+ + H_- + 8 \frac{t_L^2 + t_R^2}{U} \sum_{\sigma\sigma'} c_{\sigma}^{\dagger} \frac{\vec{\tau}_{\sigma\sigma'}}{2} c_{\sigma'^{\prime}+} \cdot \vec{S}$$

- One channel only
Goldhaber-Gordon/Oreg proposal

- Charge transfert between leads L/R and island I is suppressed below $E_I^I$
- Tough contraint : $\Delta E_I \ll T \ll E_I^I$
Experimental observation


- some evidence for scaling
- issues in the absence of complete quantitative comparison
- is there a better system?