Exotic Kondo effects in nanostructures

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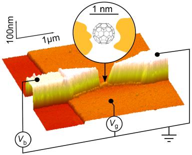
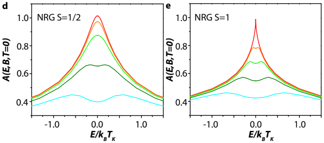
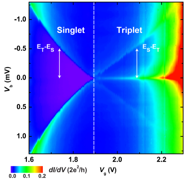






Some grateful acknowledgments

Theory side:

- A. Georges and O. Parcollet
- T. Costi

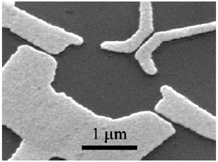
Experimental side:

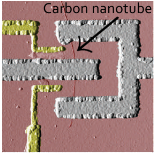
- NanoSpintronics team @ Néel Institute
  F. Balestro, V. Bouchiat, N. Roch and W. Wernsdorfer

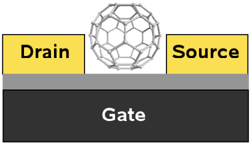
Summary

- Various quantum dot setups
- Fully screened Kondo: precise test of universality
- Under screened Kondo:
  - logarithmic approach to strong coupling
  - quantum phase transition
- Over screened Kondo: non-Fermi liquid fixed point

Various quantum dot systems

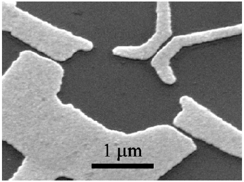


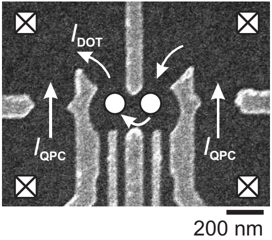


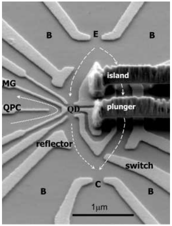


Semiconducting quantum dots

- System: 2D electron gas
- ++ great tunability and scalability by electric gates
- −− small charging energy:
  - $U = 10K$ for spin qbit experiments
  - $U = 1K$ only for Kondo experiments $\Longrightarrow$ tough!

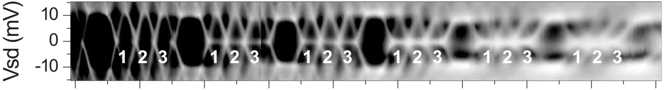


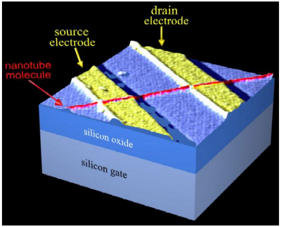


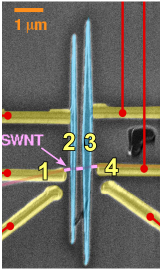


Carbon nanotube quantum dots

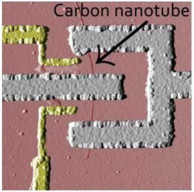
- System: long carbon molecule with gates on top
- ++ tunable & hydrid: metal, ferro or superconducting leads
- + intermediate charging energy $U = 10K - 100K$
- $\pm$ orbital degeneracy: SU(4) vs SU(2) physics







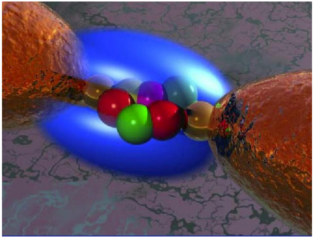
[Kontos (Paris)]

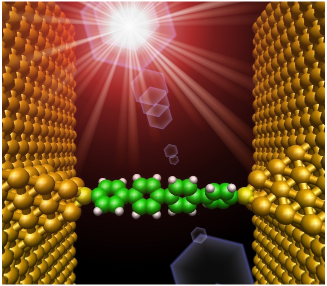


[Wernsdorfer (Grenoble)]

Molecular devices

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

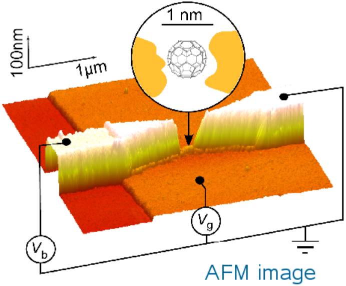


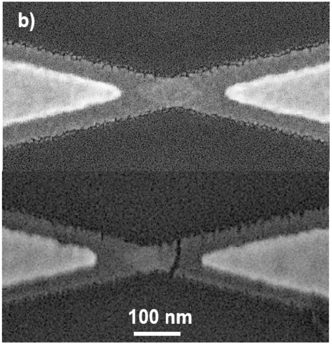


Molecular electronics HOWTO: transistor

Basic idea:

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

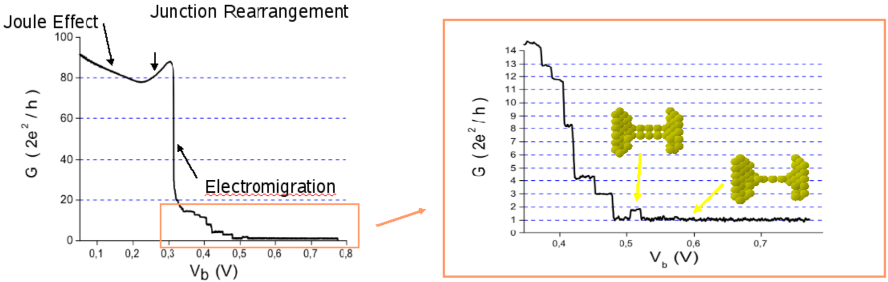




Molecular electronics HOWTO: electromigration

Recipe:

- slow voltage ramp
- interrupt the process at the right time
- hope to catch a molecule in the nanogap!

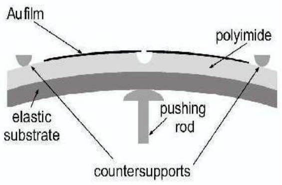


- Method development: [Park *et al.* APL (1999)]
- First ultra-low temperature measurements [Roch @ Grenoble (2008)]

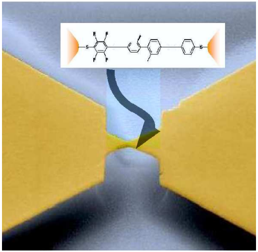
Molecular electronics HOWTO2: break junctions

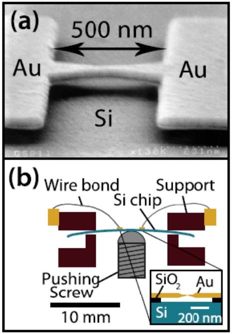
Basic idea:

- Solution: open mechanically a metal junction
- No back gate, but strain changes tunnel couplings



J. v. Ruitenbeek, Leiden '95



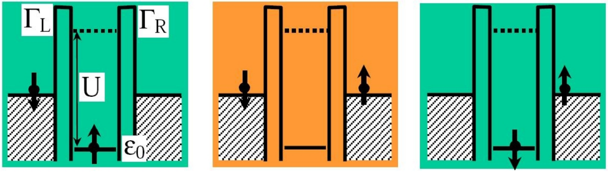


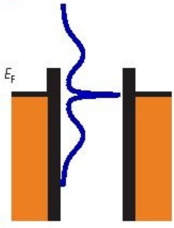
[D. Ralph (Cornell)]

Fully screened Kondo effect: universality tests

Physical origin of Kondo effect

- Lifting of a degeneracy by a Fermi sea



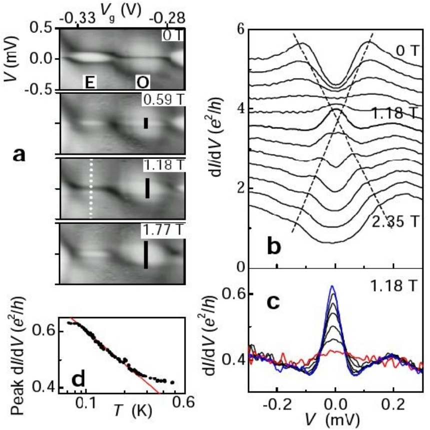


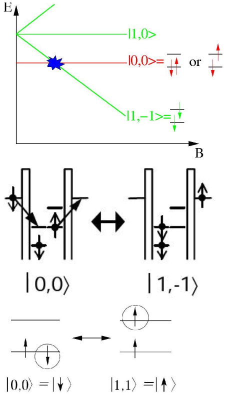
- Resonant binding of electrons near Fermi level
- Open channel for transport

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

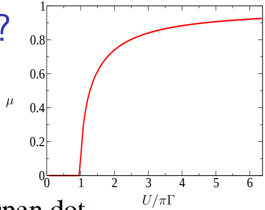
Even charge CNT quantum dot: [Nygard *et al.*, Nature (2000)]

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



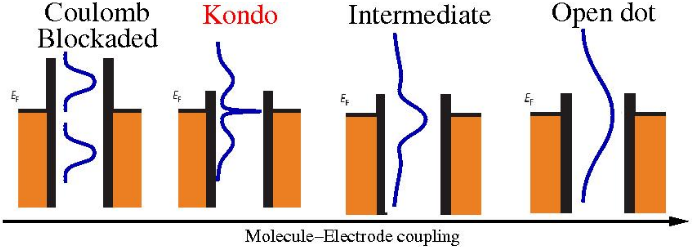


How to reach (universal) Kondo regime?



Universal (sharp) Kondo resonance requires:

- well-formed local moment i.e. large U/Γ



Consequence: only in the Kondo regime

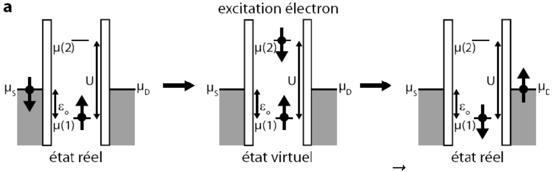
- single parameter scaling of physical quantities
  Zero-bias Conductance: $G(T) = G_0 * f(T/T_K) + G_1$

Question for experiments: how to quantify the universal regime?

Kondo model

Schrieffer-Wolff transformation: keep only spin states at $U \gg \Gamma$

$\Rightarrow$ Antiferromagnetic coupling $J_K = 8\frac{t^2}{U}$ to the Fermi sea



$$H = \sum_{k\sigma} \epsilon_k c^\dagger_{k\sigma} c_{k\sigma} + J\vec{S}\cdot\sum_{\sigma\sigma'} c^\dagger_\sigma(0)\frac{\vec{\tau}_{\sigma\sigma'}}{2}c_{\sigma'}(0)$$

High temperature limit: local moment

- $S(T) = \log 2$ and $\chi_{imp}(T) = \frac{1}{4T}$
- $G(T) = 0$

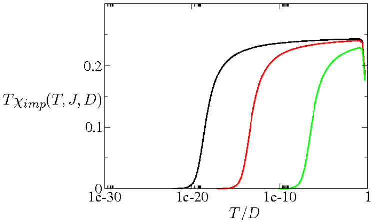
Low temperature limit: singlet state (screening)

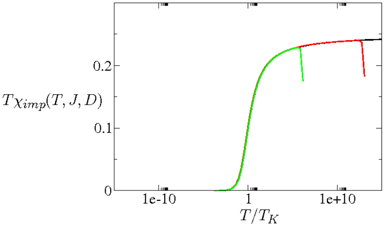
- $S(T) = 0$ and $\chi_{imp}(T) \propto \frac{1}{T_K}$
- $G(T) = \frac{2e^2}{h}$

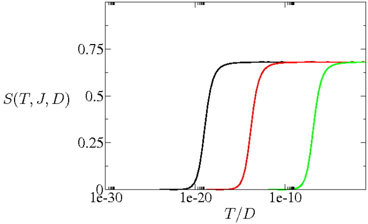
Intuitive illustration of the universal crossover

Curie constant and entropy from NRG calculations:

- Three large values of U/Γ







Perfect data collapse

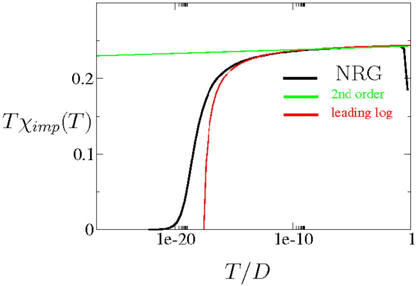
Kondo logarithms

Perturbation in $j = J/D$: badly convergent at $T \ll D$!!

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

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

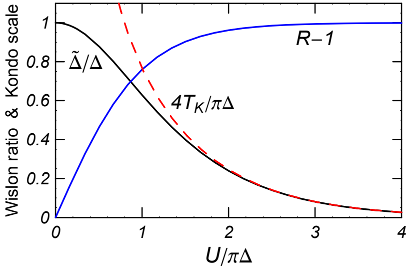
- Renormalized coupling: $j_R(T) = \frac{1}{\log(T/T_K)}$
- Kondo temperature: $T_K = De^{-D/J} = De^{-\pi U/8\Gamma}$
- Renormalized perturbation: not too useful in practice...



Precise condition to be universal

<u>Wilson ratio:</u> $R = \frac{\chi}{\gamma} * \frac{\gamma}{\chi}_{|U=0}$

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



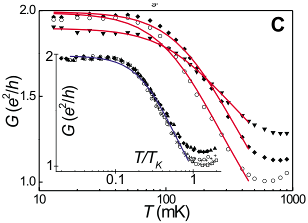
<u>Effective width:</u> $\tilde{\Delta} \equiv \tilde{\Gamma}$

<u>Kondo scale:</u> $T_K \propto e^{-\pi U/8\Gamma}$

- Small deviation to universality: $U \gtrsim 2\pi\Gamma \implies T_K \lesssim U/40$
- Similar constraints on level spacing

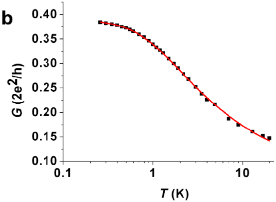
Testing the theory with experiments: $G(T)$

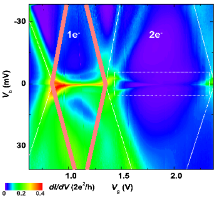
Semiconducting quantum dots: [van der Wiel *et al.* Nature (2000)]



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

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





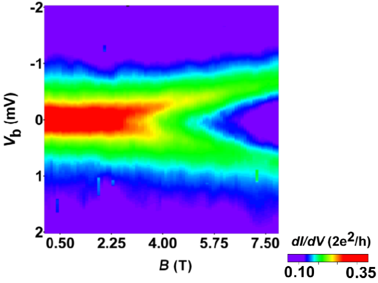
$T_K = 4K, U > 600K$ conditions are met!

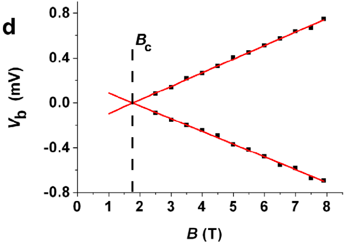
Log tails at $T \gg T_K$?

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

Magneto-transport $G(B, T)$: theory exists (NRG by Costi)

Kondo resonance splits at $T_K/2$





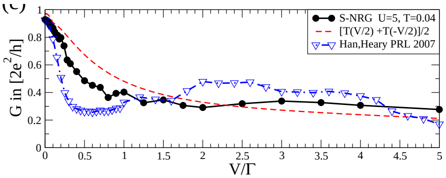
Fit of the Zeeman splitting:

- Kondo temperature $T_K = 2(g\mu_B/k_B)B_c = 4.8K$, OK!
- no quantitative comparison yet

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

Finite-bias conductance

- Equilibrium case: $\Gamma_L \gg \Gamma_R$ [i.e. $G(T=0) \ll 2e^2/h$]
  $\Longrightarrow$ spectroscopy of the Kondo resonance
- Out-of-equilibrium case: $\Gamma_L \simeq \Gamma_R$ [i.e. $G(T=0) \simeq 2e^2/h$]
  - Main physical effect: enhanced scattering from large current density kills the Kondo resonance
  - Crucial and tough problem for many-body theory
  - Reliable experimental data badly needed!
- Recent non-equilibrium NRG and QMC simulations:
  [Anders PRL (2009), Han PRL (2007)]



Checking Fermi Liquid Theory

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

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

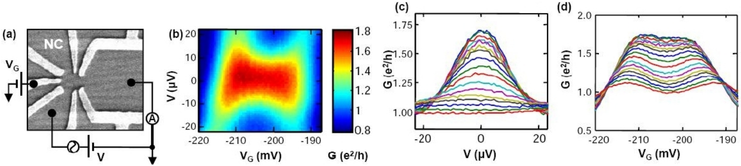
Out of equilibrium Fermi Liquid theory: [Oguri JPS (2004)]

$$\begin{aligned} \alpha \equiv \frac{c_V}{\pi^2 c_T} &= \frac{3}{4\pi^2} \frac{1+5(R-1)^2}{1+2(R-1)^2} \\ &= \frac{3}{2\pi^2} \simeq 0.16 \, [\text{Kondo regime} \, (R=2)] \\ &= \frac{3}{4\pi^2} \simeq 0.08 \, [\text{uncorrelated} \, (R=1)] \end{aligned}$$

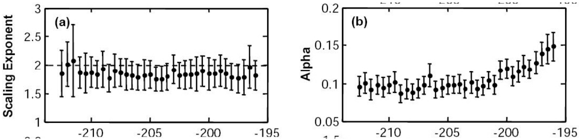
Note: α depends on Γ_L/Γ_R in general
but not any more in the $R=2$ limit (universality again!)

Hunt for α: 2DEG quantum dot data

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



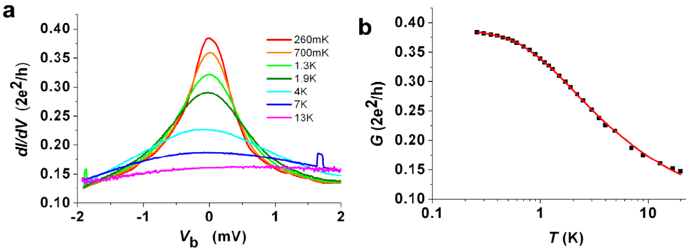
Fermi liquid coefficients:



- Nice systematics!
- $0.8 < \alpha < 1.6$: signature of intermediate correlations?!

Hunt for α: molecular quantum dot data

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



- Here $T_K = 4K \implies$ less noise!
- For this only sample: one extracts $\alpha \simeq 1.5$, OK!
- More studies needed (several T_K and lead asymmetries)

Under screened Kondo effect: logarithmic singularities

Exotic Kondo: historical note

Seminal work: Nozières-Blandin (1981)

- Initial motivation: realistic aspects of Kondo alloys!
- They found surprising physics...

Flurry of theory work since then:

- Under and Over screened Kondo (this talk)
- Kondo in a superconductor (very active experimentally)
- SU(4) Kondo (studied in carbon nanotubes)
- Two impurity Kondo
- Kondo in a pseudogap metal
- Charge or orbital Kondo
- Role of phonons
- Kondo in Luttinger liquids
- ...

Exotic Kondo: experimental realisations?

The challenge in condensed matter:

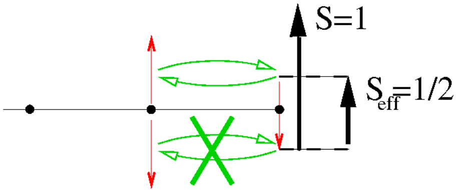
- $++$ Bulk measurement
  singular magnetic response or extensive entropy easy to spot
- $-$ little tunability
- $--$ conspiracy theory: always a Fermi liquid?? (see later)
- New system: $Si_{1-x}Fe_x$ may be underscreened ??

The challenge in quantum dots:

- $++$ Tunability and man-designed Hamiltonian
- $\pm$ Single impurity measurement
- $--$ Only conductance available
  $\Longrightarrow$ requires very careful and tough measurement

Under screening: Nozières-Blandin argument

Spin $S = 1$ and single screening channel:



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

$\Longrightarrow$ scale dependence: $J_{eff}(T) \propto \frac{1}{\log(T/T_K)}$ at $T \ll T_K$

$\Longrightarrow$ $J_{eff}(T)$ vanishes at low T

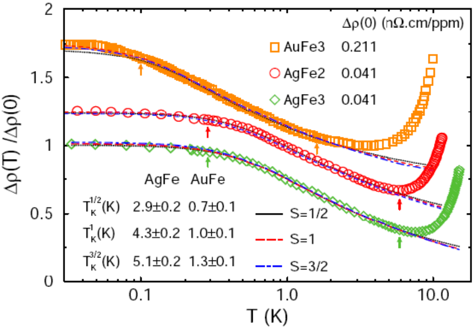
Transport: logarithmic approach to unitarity

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

Disgression: Kondo anomalies in wires

Resistance of $Ag_{1-x}Fe_x$ and $Au_{1-x}Fe_x$: [Costi *et al.* PRL (2009)]

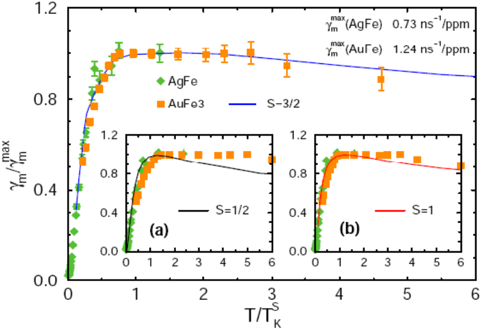
- LDA predicts spin $S = 3/2$ for Fe and 3 orbitals involved in screening process



- Data compatible with full screening (no underscreening!)
- Theory cannot distinguish the spin value S

Disgression: Kondo anomalies in wires

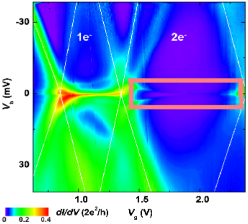
Inelastic contribution to the resistivity:

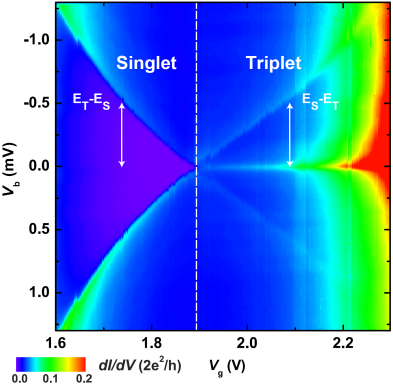


- This allows to discriminate the spin value:
  $\Longrightarrow S = 3/2$ and 3 orbital involved!

Molecular transistor: even charge

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

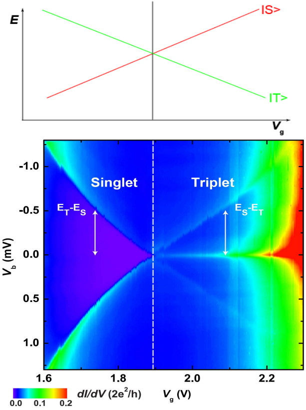


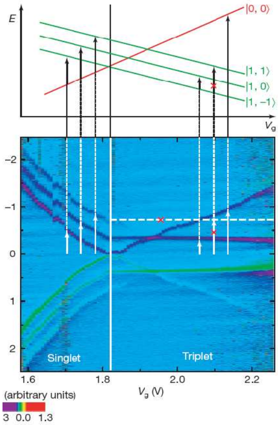


Life and death of a spin $S = 1$ Kondo anomaly:

- change in the magnetic ground state of the quantum dot?

Identifying the spin states: gate voltage scan at $B = 3T$



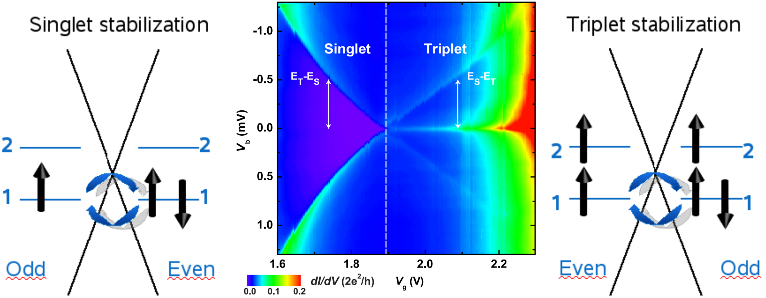


- Zeeman effect agrees with spin 0 or spin 1 ground state
- Gate-induced magnetic splitting (tunable Hund's rule!)

Origin for the gating effect

Role of the leads: energy gain by charge fluctuations

Hopping from level 1 or 2 $\Longrightarrow \delta E_{1,2} = -\frac{t_{1,2}^2}{E_{add}}$



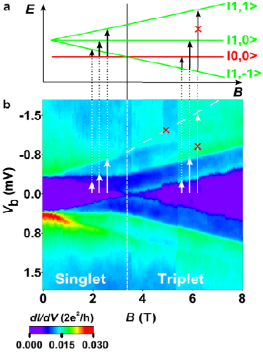
Conclusion: hopping asymmetry $t_1 \gg t_2$

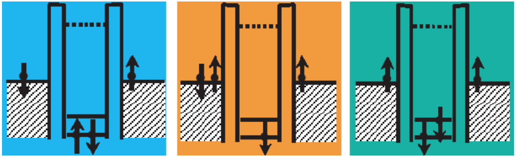
Crucial observation: single screening channel!!

Second evidence for single screening channel

Magnetic field effect:

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

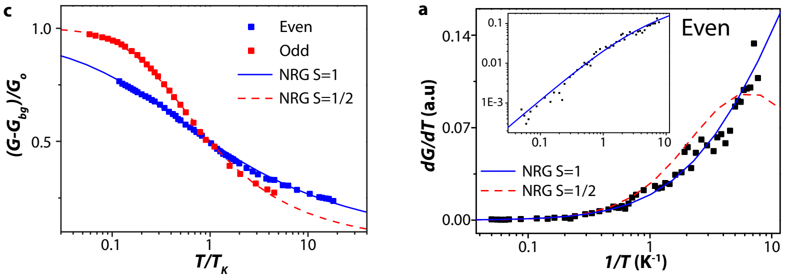




- Kondo coupling: $J_K \propto \frac{t_1 t_2}{E_c}$ small!

Testing the underscreened Kondo scenario

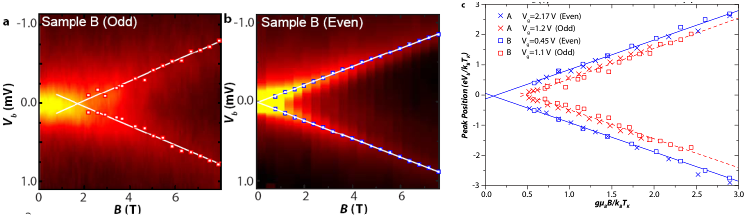
Analysis of the spin $S = 1$ Kondo anomaly: [Roch *et al.*, PRL (2009)]



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

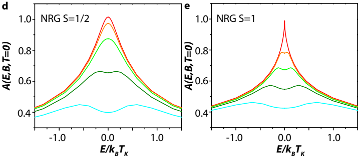
Magnetic field effect: smoking gun?

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



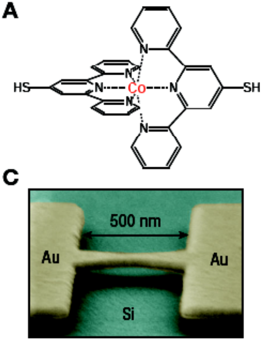
- Splitting occurs at lower magnetic fields for $S = 1$!

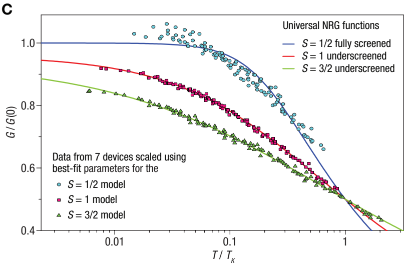
NRG calculations: $S = 1$ Kondo resonance very sensitive to field



Break junction device

Cobalt in a "cage": [Parks *et al.* cond-mat (2010)]

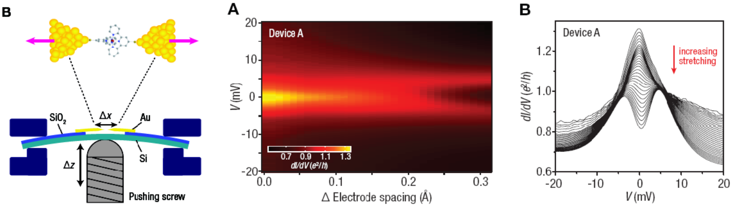




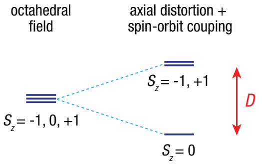
Underscreened Kondo anomaly: clearly logarithmic below T_K

Unexpected effect

Stretching the molecule:



Interpretation: strain induced magnetic anisotropy



Putting the $|1, 0\rangle$ state down: kills underscreened Kondo

Under screened Kondo effect: quantum phase transition

Some general arguments

Quantum phase transition:

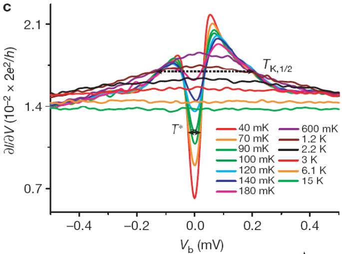
- continuous change between two physically different ground states
- purely zero temperature phenomenon with observable consequences at finite temperature

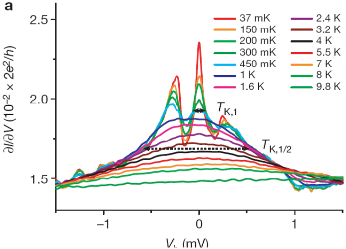
Underscreened Kondo:

- a complex many-body ground state (spin+contacts)
- remanent entropy: analogous to a disordered phase
- sensitive to perturbation towards zero entropy state
- zero temperature change of ground state degeneracy: (impurity) quantum phase transition (not a level crossing)

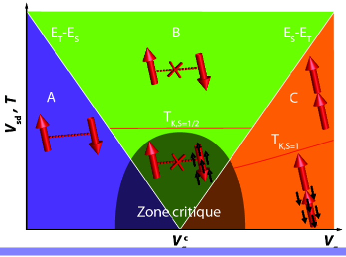
Back to Grenoble experiment

Around the singlet-triplet crossing:

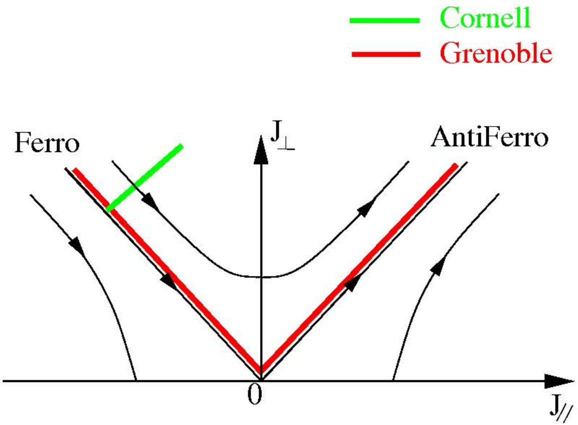




Interpretation:



Where are we in parameter space?



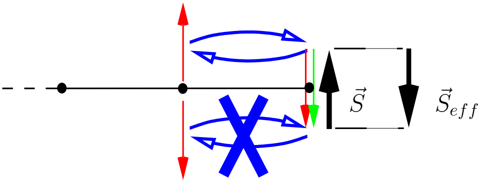
Over screened Kondo effect:
Non Fermi Liquid

The model and Nozières-Blandin argument

Two independent Fermi seas (m=1,2):

$$H = H_1 + H_2 + \sum_{\sigma\sigma'} \sum_m J_m \, c^\dagger_{\sigma m} \frac{\vec{\tau}_{\sigma\sigma'}}{2} c_{\sigma' m} \cdot \vec{S}$$

Strong coupling argument at $J_1 = J_2$ large



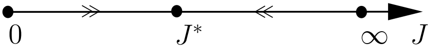
- Effective spin $S_{eff} = 1/2$ (residual entropy)
- Effective Kondo coupling $J_{eff} \propto +t^2/J$: antiferromagnetic!
  $\Longrightarrow$ strong coupling fixed point is unstable!

Intermediate coupling fixed point

Weak coupling RG: next order

$$\Lambda \frac{dJ}{d\Lambda} = -J^2 + KJ^3$$

- $K = \#$ channels ($K = 2$ or more)
- $J^* = \frac{1}{K}$ NFL fixed point controlled if $K \gg 1$
- Break down of quasiparticle picture!



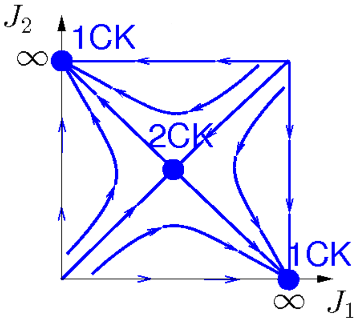
What's known: (NRG, CFT, Bosonization...)

- $S(T=0) = \log(\sqrt{2})$: weird!
- $\chi(T) \propto \log(T_K/T)$
- $G(T) = G_0[1 - a\sqrt{T/T_K}]$ at $T \ll T_K$

Channel anisotropy kills 2CK

Weak coupling RG: with $J_1 \neq J_2$

$$H = H_1 + H_2 + \sum_{\sigma\sigma'} \sum_m J_m \, c^\dagger_{\sigma m} \frac{\vec{\tau}_{\sigma\sigma'}}{2} c_{\sigma' m} \cdot \vec{S}$$

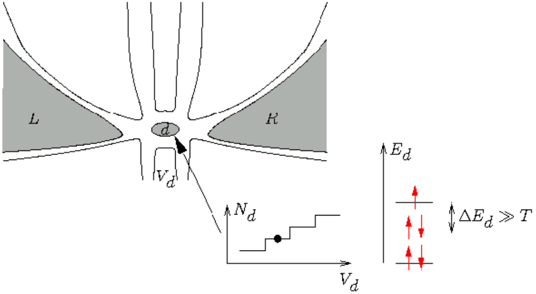


- $\Longrightarrow$ fine tuning needed!
- but can one realize such Hamiltonian with quantum dots?

Why 2 leads experiments give only 1 channel?

Kondo Hamiltonian: $\alpha = L, R$

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

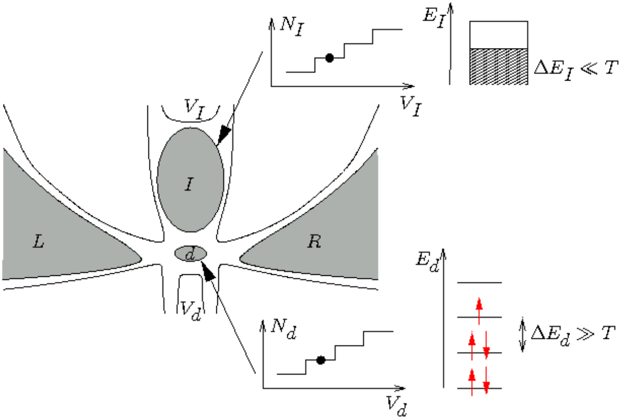


Matrix of couplings: $J_{\alpha,\alpha'} \propto \frac{t_\alpha t'_\alpha}{U}$ has only one non-zero eigenvalue

$$\Longrightarrow H = H_+ + H_- + 8\frac{t_L^2 + t_R^2}{U} \sum_{\sigma\sigma'} c^\dagger_{\sigma+} \frac{\vec{\tau}_{\sigma\sigma'}}{2} c_{\sigma'+} \; \cdot \; \vec{S}$$

- One channel only

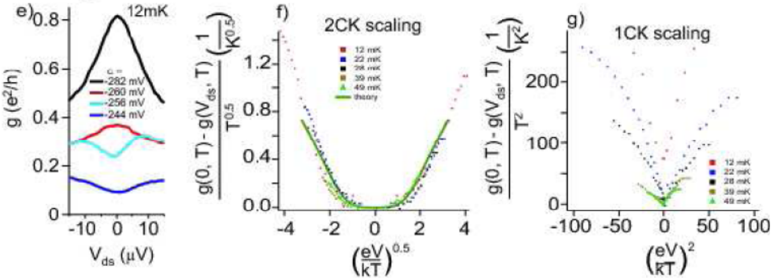
Goldhaber-Gordon/Oreg proposal



- Charge transfert between leads L/R and island I is suppressed below E_c^{I}
- Tough contraint: $\Delta E_{\mathrm{I}} \ll T \ll E_c^{\mathrm{I}}$

Experimental observation

[Potok *et al.* Nature (2007)]



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

Conclusion

Conclusion

- Beyond the standard (one channel $S = 1/2$) Kondo effect:
  - rich physics brought by increased complexity
  - great potentialities in molecular electronics
- Theory is ripe to handle complicated impurity problems
  - not yet out-of-equilibrium
  - not yet with ab-initio
- First observations of under and over screened situations made only recently
- Other effects to look for: two-impurity Kondo (and others)