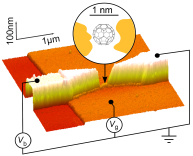
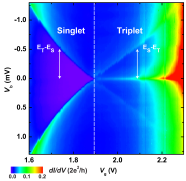
Singlet-triplet transition in a single molecule transistor

N. Roch, S. Florens, V. Bouchiat, W. Wernsdorfer and F. Balestro

Néel Institute - CNRS Grenoble

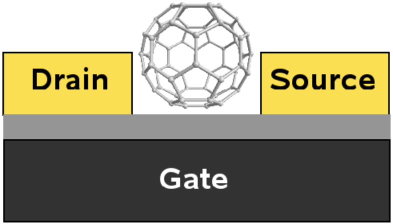




Summary

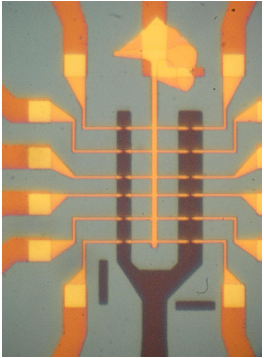
- Sample fabrication: electromigration and measurement setup
- Odd diamond: spin 1/2 Kondo effect
- Even diamond: identifying the magnetic states
- Even diamond: analysis of various Kondo effects
- Even diamond: singlet-triplet transition
- Outlook: conclusion and perspectives

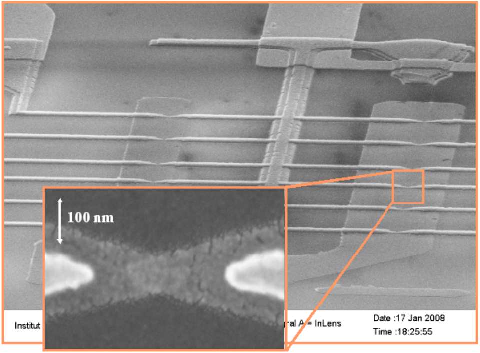
Sample fabrication



First step: lithography

Gold nanowires+Al/Al_2O_3 backgate



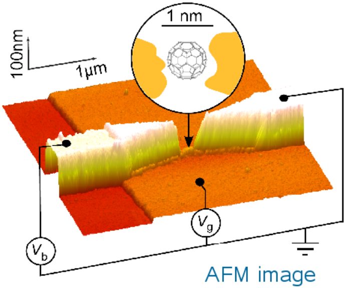


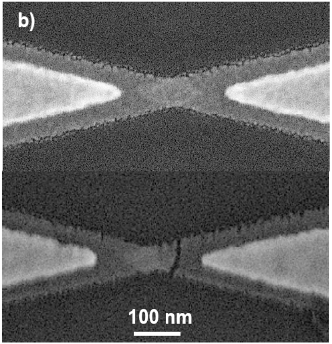
Even with E-Beam Lithography, it is not possible to reach the single molecule size !

Single molecule transistor

Basic idea:

- coat sample with C_{60} molecules
- break the junction via electromigration
- repeat until success

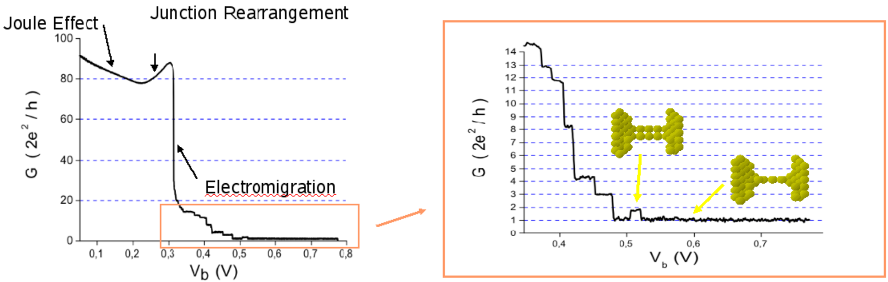




Second step: electromigration

Important recipe:

- slow voltage ramp
- interrupt the process quickly and at the right time



- Method development: Park *et al.* (1999)
- With C_{60}: Lu and Natelson (2004), Parks *et al.* (2007),...
- Other molecules: Liang *et al.* (2002), Osorio *et al.* (2007),...

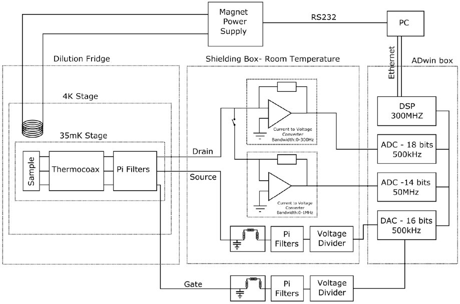
Measurement setup

Electromigration stage:

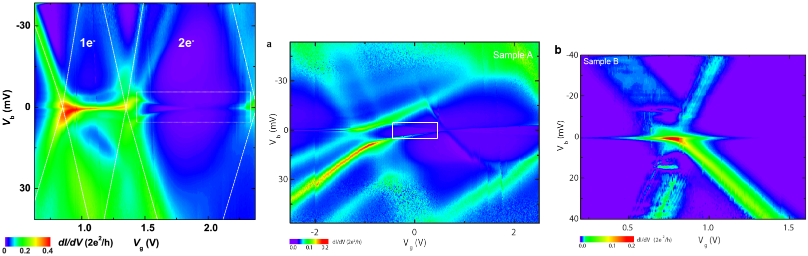
- Low series resistance
- Fast feedback loop

Crucial aspects:

- Dilution fridge ($35\text{mK} < T < 20\text{K}$)
- Low temperature filtering
- Local electrostatic gate (thin oxide layer)



Several samples: typical conductance plots at 35mK

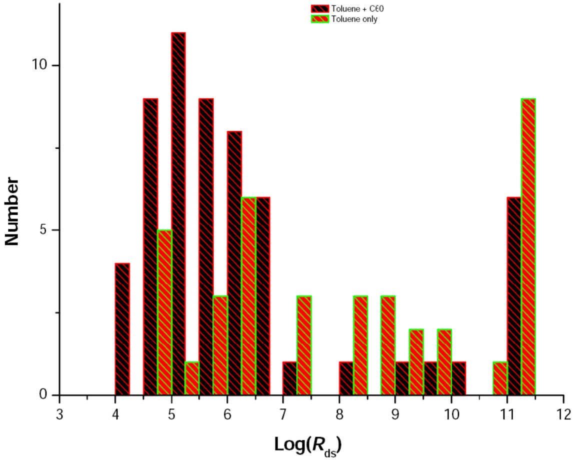


Intermediate coupling: Coulomb blockade and Kondo anomalies

Problem: Lack of reproducibility

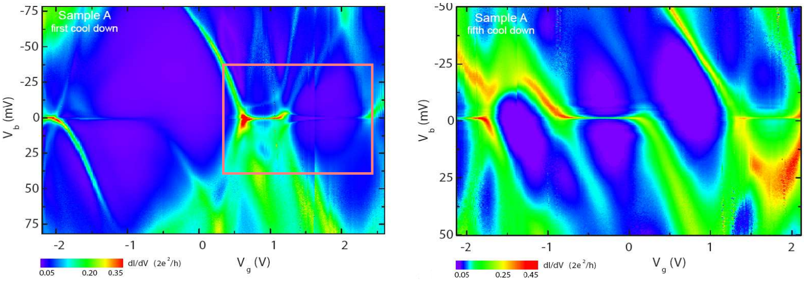
What's in there?

Statistical analysis of resistance: gold nanoparticules vs. C_{60}?



Coulomb diamonds

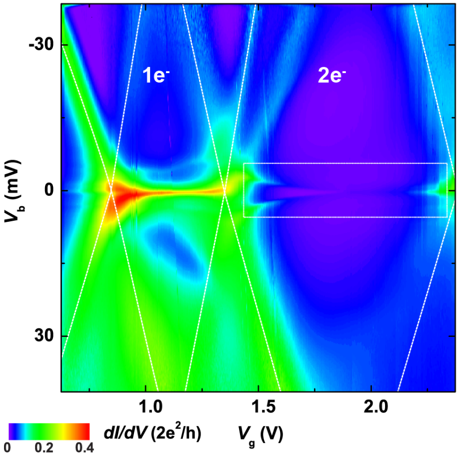
Two scans after thermal cycling:



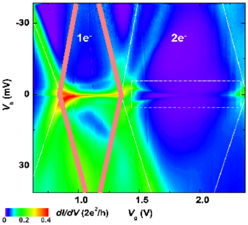
<u>Low charging energy:</u> important screening effects?

<u>Similar observations:</u> Kubatkin *et al.* (2003), Osorio *et al.* (2007).

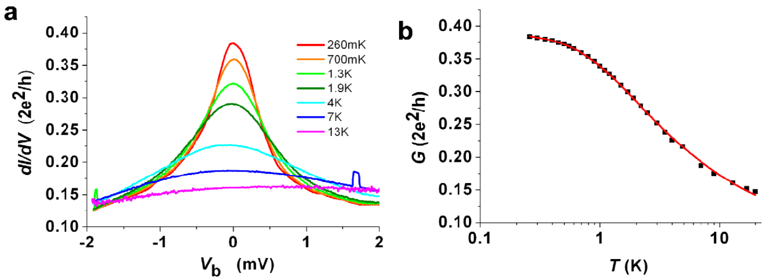
"Best sample": global view on the transport data



Odd charge Coulomb diamond



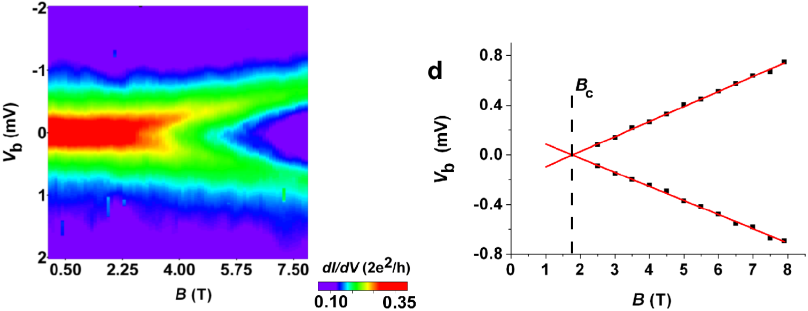
Conductance: spin 1/2 Kondo effect



HWHM of $dI/dV(V_b)$: Kondo temperature $T_K = 4.4\text{K}$

NRG fit of $G(T)$: Kondo temperature $T_K = 4.5\text{K}$

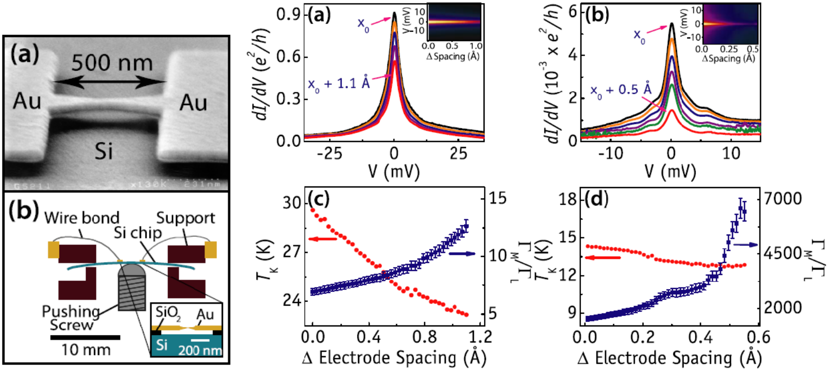
Magnetic field dependence



Fit of the Zeeman splitting:

- g-factor = 2
- Kondo temperature $T_K = 2g\mu_B B_c / k_B = 4.8\text{K}$

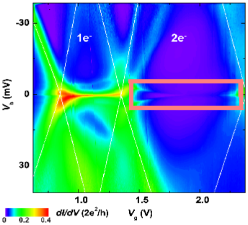
Comparison with previous C_{60} data



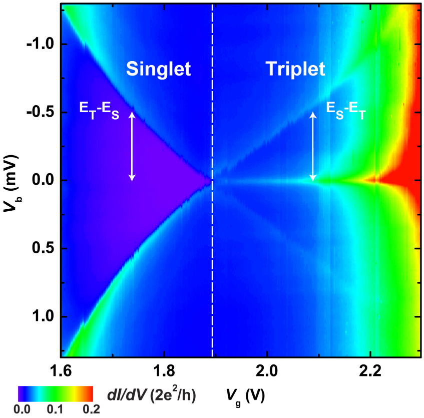
Data from Parks et al. (2007).

Wide range of Kondo temperatures: 10K$<T_K<$60K

Even charge Coulomb diamond



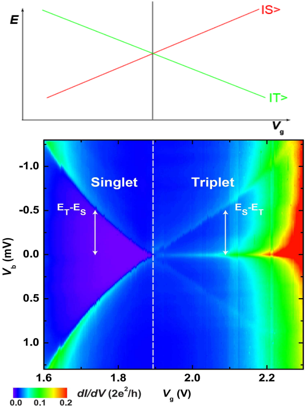
Gate-induced level crossing at zero magnetic field

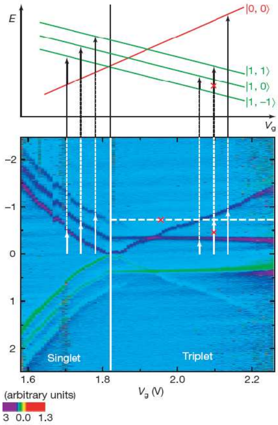


Life and death of a Kondo anomaly:

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

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

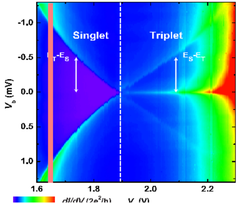


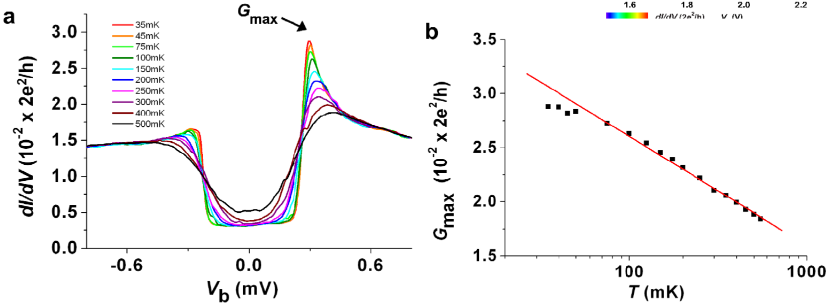


- Zeeman effect agrees with spin 0 or spin 1 ground state
- Gate-induced magnetic splitting

Singlet side

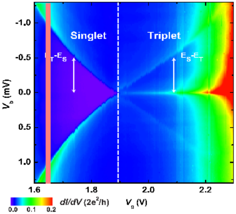
Singlet-side: non-equilibrium Kondo

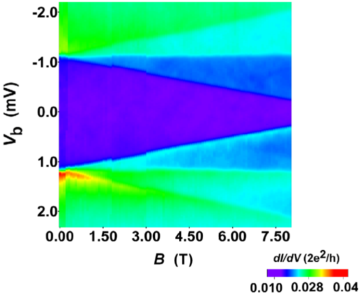


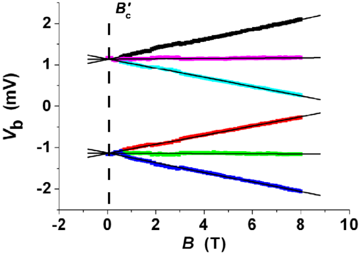


Similar data in carbon nanotubes by Paaske et al. (2006)

Singlet-side: magnetic field-dependence

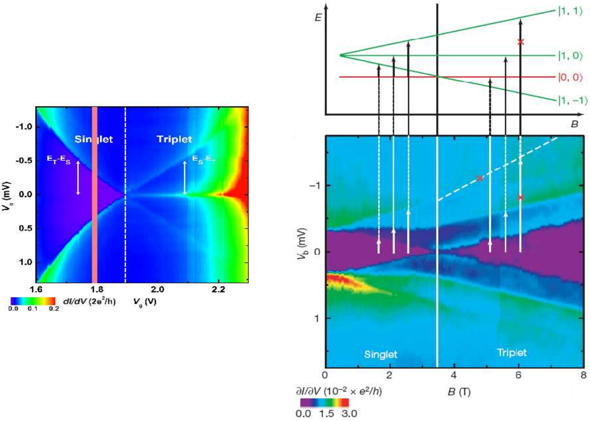






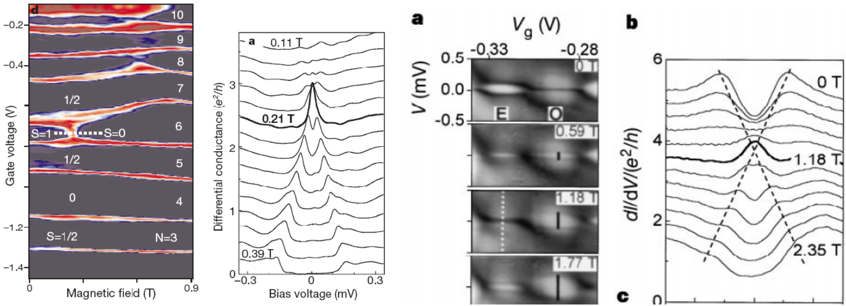
Low field splitting of the Kondo peak: $B'_c < 50m$T. Theory?

Magnetic field spectroscopy: field tuning at fixed V_g



Crossing of singlet and lowest triplet: no Kondo enhancement!

Comparison: Zeeman induced singlet-triplet transition



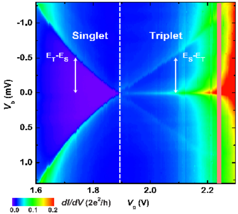
Data from Sasaki *et al.* and Nygard *et al.* (2000).

No ZBA for C_{60}: the two orbitals tunnel differently?!

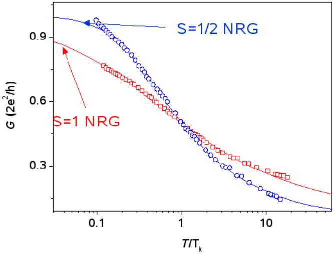
Triplet side

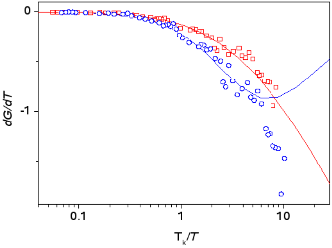
Zero-bias anomaly: $S = 1$ underscreened Kondo?

Rescaling of the data: $G(T) \rightarrow G_{bg} + \alpha G(\frac{T}{T_K})$



Comparison to NRG: Costi (2000)

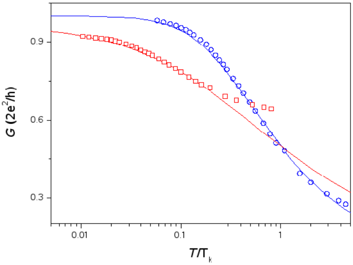


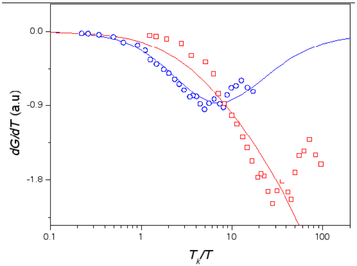


Best evidence for spin $S = 1$ Kondo: logarithmic all the way

Comparison to odd Coulomb diamond data

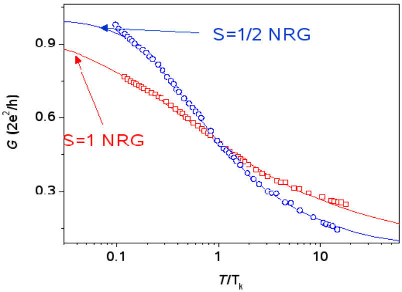
NRG: $S = 1/2$ and $S = 1$ calculations vs. data:

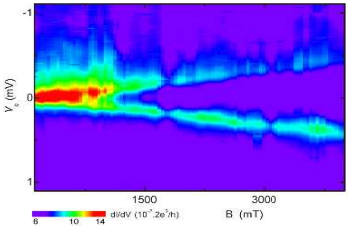


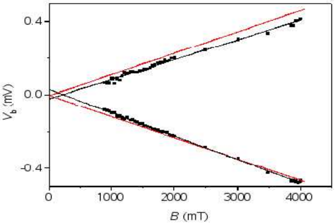


Fermi Liquid upturn incompatible with $S = 1$ NRG data

Zeeman effect on the $S = 1$ Kondo anomaly





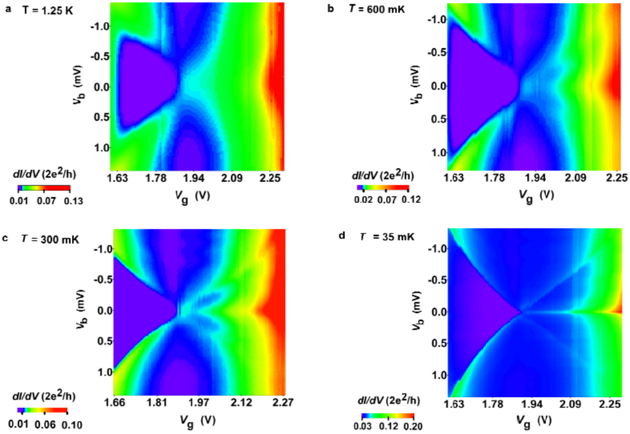


NRG fit of G(T): $T_K = 1.1\text{K}$

Magnetic field splitting: $T_K = 500\text{mK}$

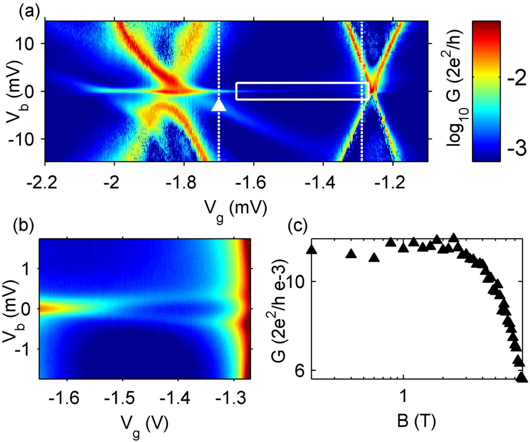
Question: are there $S = 1$ NRG data for $\rho(\omega, B)$?

Temperature dependence: revealing the $S = 1$ Kondo peak



At high temperature all states are mixed up!

Comparison to previous data

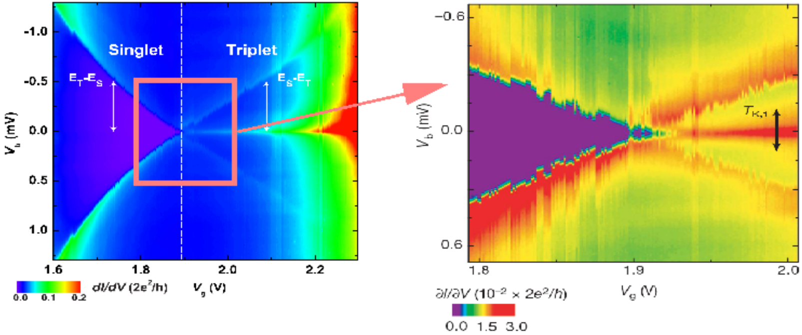


Carbon nanotube data from Quay *et al.* (2007)

Difference: thermal smearing of the transition (noise?)

Close to the singlet-triplet transition

A closer look on the transition point



All magnetic excitations merge together

Nature of the transition

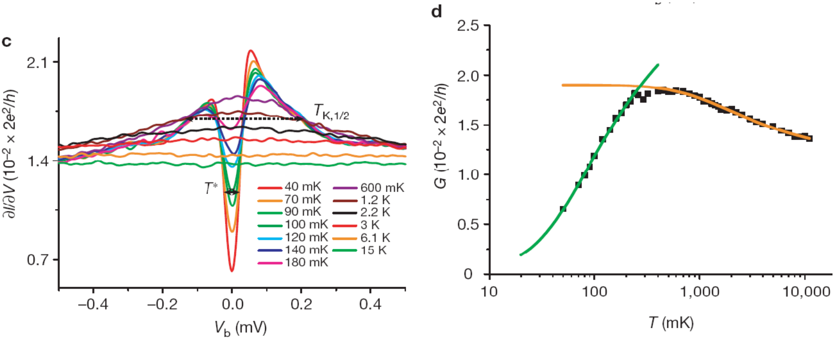
Several theoretical studies:

- NRG: Vojta and Bulla (2002), Hofstetter and Schoeller (2002)
- Scaling theory: Glazman and Pustilnik (2001), Chung, Zarand and Woelfle (2007)
- ...

Physical picture:

- Singlet and triplet states dissociate at the transition
- One orbital gets fully screened
- On the singlet side: the second orbital binds via a second stage Kondo effect

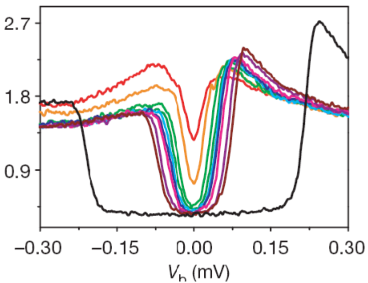
Singlet side: two-stage Kondo

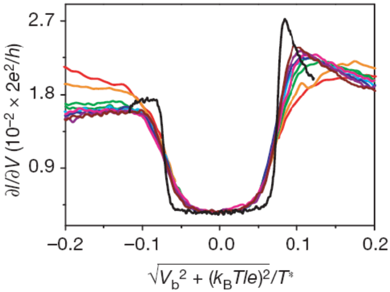


- $T_{K,1/2}$: single spin Kondo temperature
- T^*: binding energy of the singlet

Singlet side: scaling

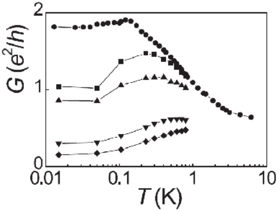
From close to far from the transition point:



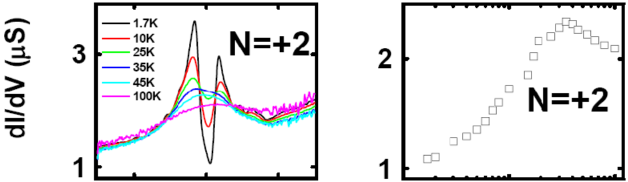


- Kondo dip well developped
- Scaling difficult because of thermal filling in the dip

Comparison to previous data

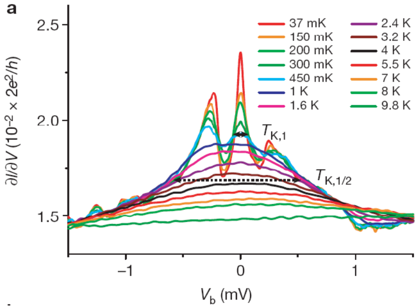


Data on GaAs quantum dots by van der Wiel *et al.* (2002)



Data on OPV5 molecule by Osorio *et al.* (2007)

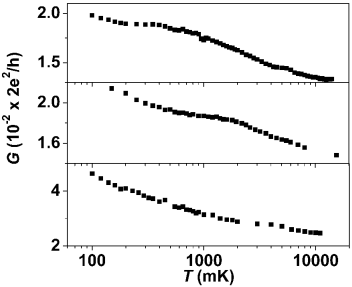
Triplet side: three energy scales

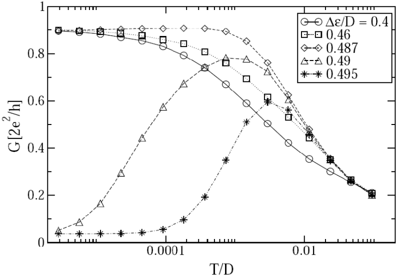


- $T_{K,1/2}$: single spin Kondo temperature
- $T^{K,1}$: spin $S = 1$ Kondo temperature
- $E_S - E_T$: Kondo sidebands

Triplet side: temperature dependence

Close to far from the transition point:



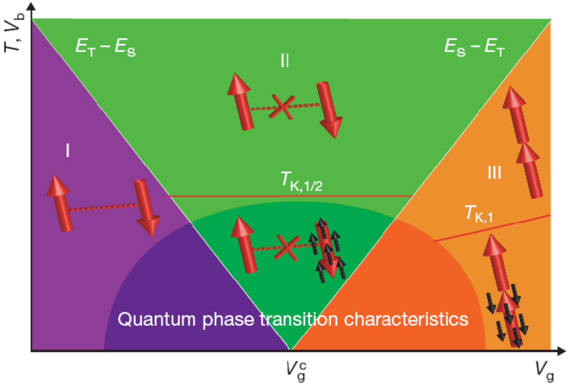


NRG by Hostetter and Schoeller (2002)

Kink in $G(T)$ at the singlet-triplet splitting clearly visible

Question: is this really compatible with NRG?

Summary


T, V_b
E_T − E_S
II
E_S − E_T
I
T_K,1/2
III
T_K,1
Quantum phase transition characteristics
V_g^c
V_g

Conclusion

Conclusion and perspectives

Summary of results:

- Very clean $s = 1/2$ Kondo effect in C_{60} quantum dot
- First attempt to study $S = 1$ underscreened Kondo effect
- Detailed investigation of singlet-triplet transition

Advantages of molecules:

- Molecular junctions: new toy to explore Kondo physics (chemistry!)
- Larger energy scales: better test-bed for theory

Future experimental goals:

- Fill C60 or explore more complex molecules
- Towards superconducting single molecule junctions