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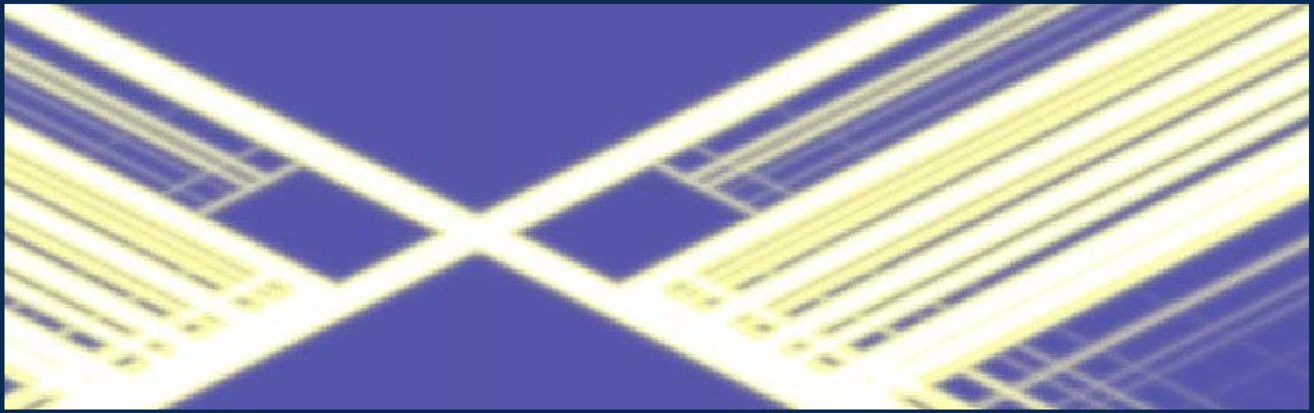
Faculty of Science, Institute for Theoretical Physics, Condensed Matter Theory

Magnetic N@C_{60} single-molecule transistors

Towards modeling of real devices

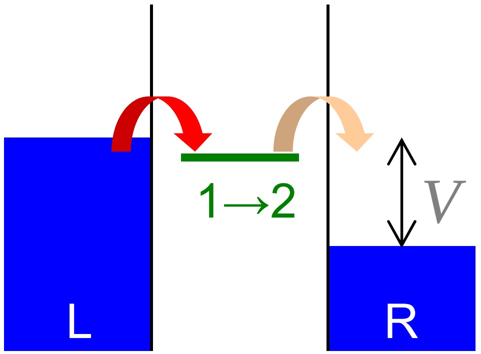
Carsten Timm

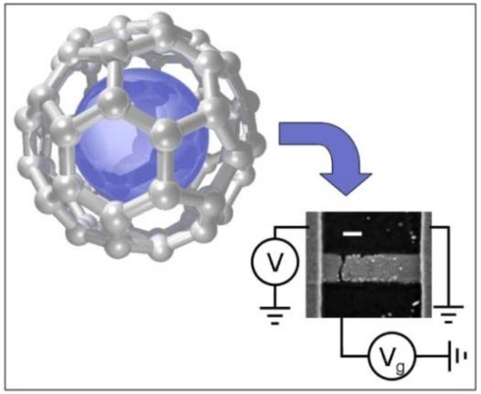


Max Bergmann Symposium 2008

Overview

- Master equation formalism
- Endohedral $N@C_{60}$
- $N@C_{60}$ transistors





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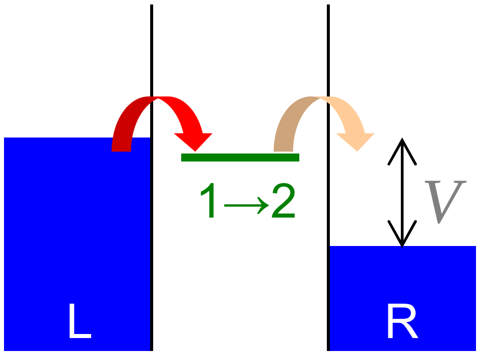
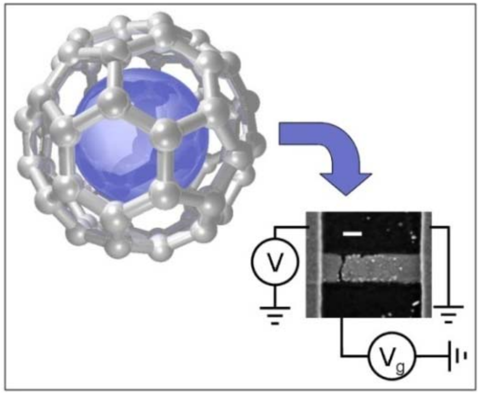
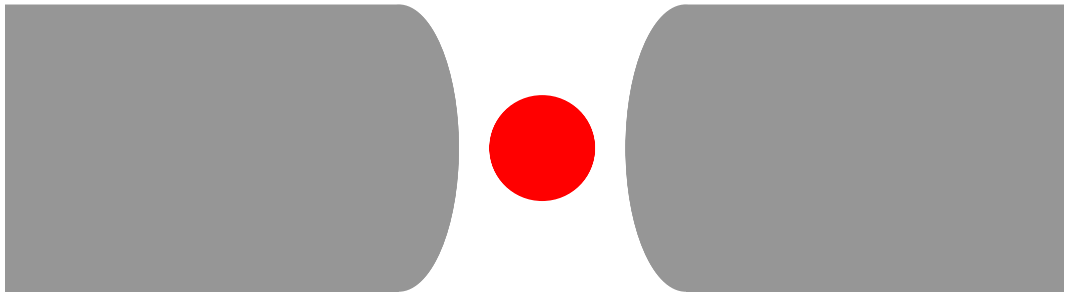




Small system coupled to large reservoirs



Here: quantum dot / molecule coupled to bulk leads

$\overline{A_{\mathrm{dot}}}(t) = $? $\overline{I}(t) = $?

dot observable current

$$\overline{A_{\mathrm{dot}}}(t) = \mathrm{Tr}\, \rho(t)\, A_{\mathrm{dot}}$$ with the density operator $$\rho(t) \cong \rho_{\mathrm{dot}}(t) \otimes \rho^0_{\mathrm{leads}}$$

Cannot solve this because H is complicated!

Now what?

$A_{\rm dot}$ only depends on the dot: $\overline{A_{\rm dot}}(t) = \mathrm{Tr}\, \rho_{\rm dot}(t)\, A_{\rm dot}$

with reduced density operator (in "small" dot Hilbert space)

$$\rho_{\rm dot} \equiv \sum_i \langle\langle i |\, \rho\, | i \rangle\rangle \equiv \mathrm{tr}_{\rm leads}\, \rho$$

basis of lead (reservoir) states only

Big question: What is the equation of motion of $\rho_{\rm dot}(t)$?

The Master Equation!

Many different approaches; all start from the von Neumann equation:

$$\frac{d\rho}{dt} = -i\,[H, \rho] \quad \Longrightarrow \quad \frac{d}{dt}\rho_{\rm dot} = -i\,\mathrm{tr}_{\rm leads}[H, \rho(t)]$$

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Wangsness-Bloch-Redfield master equation

Hamiltonian $H = H_{\text{dot}} + H_{\text{leads}} + H_{\text{hop}}$ ← here: electron hopping between dot and leads

- iterate von Neumann equation to expand to second order in H_{hop}
- assume product state with leads in equilibrium at time t: $\rho(t) \cong \rho_{\text{dot}}(t) \otimes \rho^0_{\text{leads}}$

  means that dot and leads are uncorrelated (strong but superfluous assumption)

$$\frac{d}{dt}\rho_{\text{dot}} \cong -i\,[H_{\text{dot}}, \rho_{\text{dot}}(t)] - \int_{-\infty}^{t} dt'\, \text{tr}_{\text{leads}} \left[H_{\text{hop}}, \left[e^{-i(H_{\text{dot}}+H_{\text{leads}})(t-t')} H_{\text{hop}} e^{i(H_{\text{dot}}+H_{\text{leads}})(t-t')}, \rho_{\text{dot}}(t) \otimes \rho^0_{\text{leads}}\right]\right]$$

Wangsness-Bloch-Redfield master equation

see C.T. , PRB **77**, 195416 (2008)

not of the form $\dfrac{d\rho_{\text{dot}}}{dt} = -i\,[\tilde{H}, \rho_{\text{dot}}]$

$\rightarrow$ time evolution not unitary, includes relaxation

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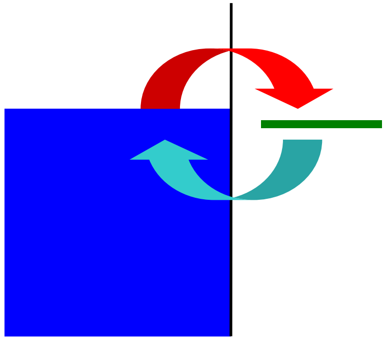
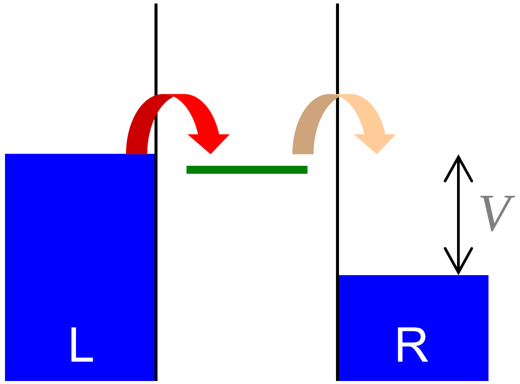
Case 1: single reservoir (particle & energy bath)

dot approaches equilibrium for $t \to \infty$:

$$\rho_{\rm dot} \propto e^{-\beta(H_{\rm dot}-\mu N_{\rm dot})}$$



Case 2: two leads in *separate* equilibrium—*e.g.* different chemical potential



Have a bias voltage V

Keeps dot out of equilibrium but approaches a steady state

Rate equations

Unperturbed dot many-particle eigenstates: $H_{\text{dot}}\,|m) = E_m\,|m)$

If off-diagonal components of ρ_{dot} in basis $\{|m)\}$ relax rapidly (rapid dephasing):

sufficient to keep only diagonal components

$P_m \equiv (m|\,\rho_{\text{dot}}\,|m)$ probabilities of dot states $|m)$

obtain rate equations $$\frac{dP_m}{dt} = \sum_n \left(R_{n\to m}P_n - R_{m\to n}P_m\right)$$

in *out*

➡ $P_m(t)$ ➡ observables, *e.g.* $I(t) \equiv \overline{I}(t)$

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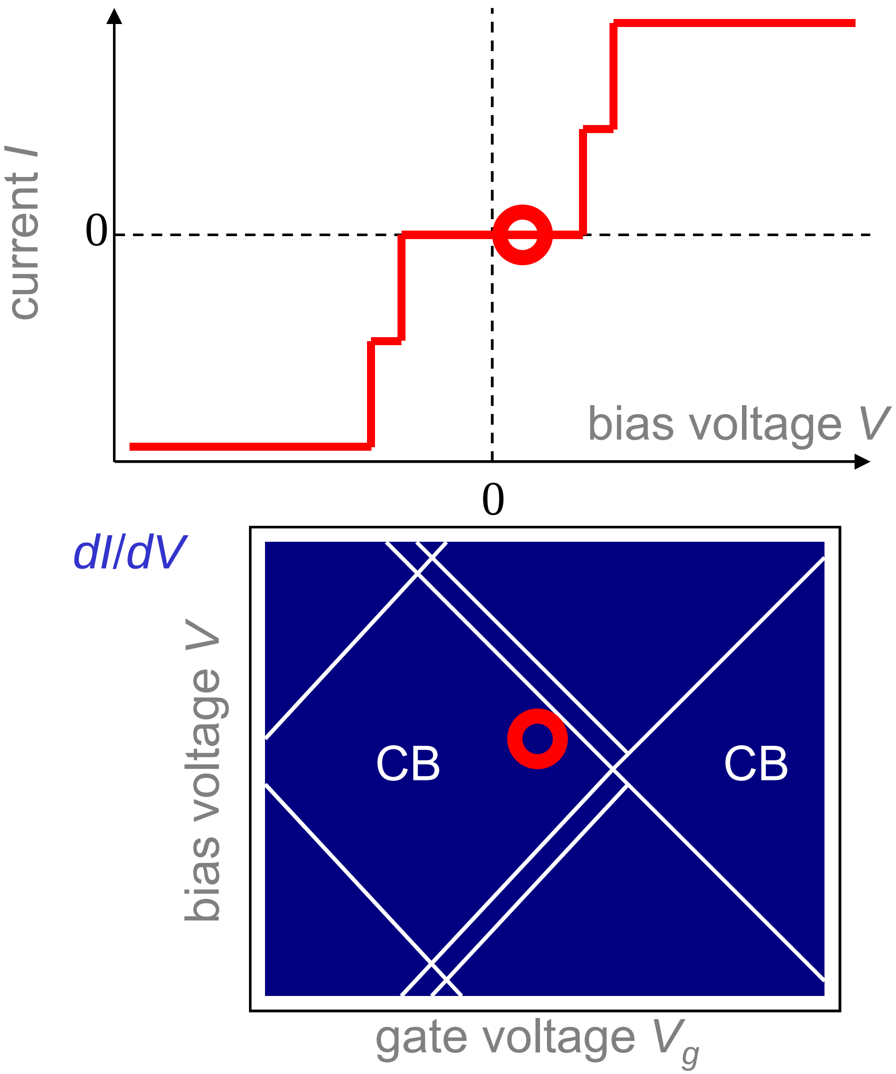
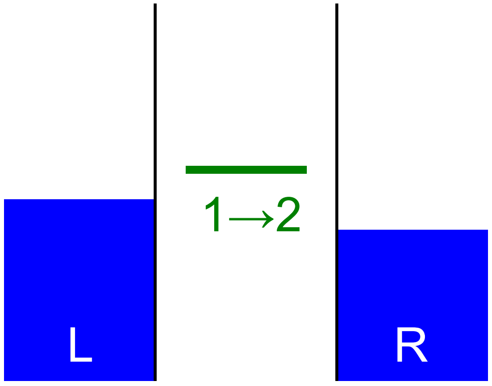
Generic behavior described by rate equations

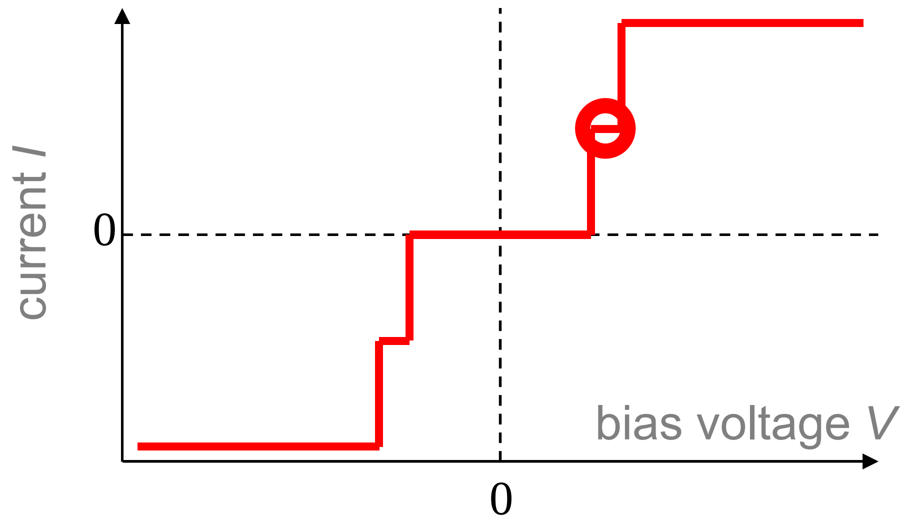


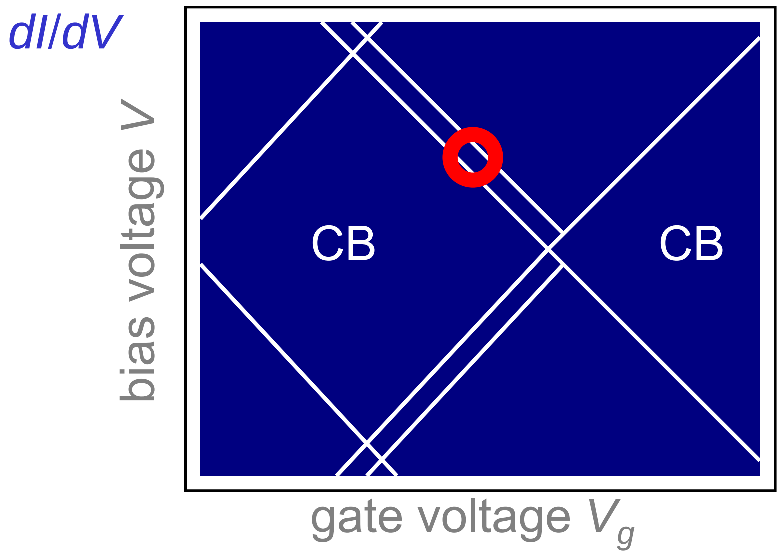


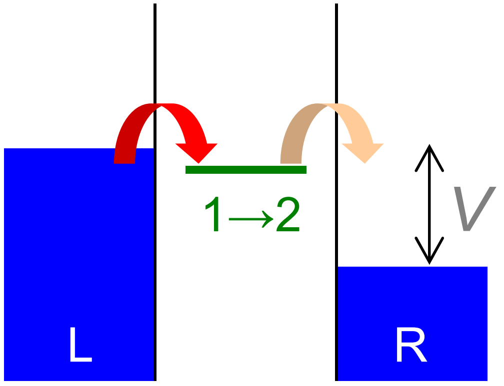
very small current:

Coulomb blockade

Generic behavior described by rate equations

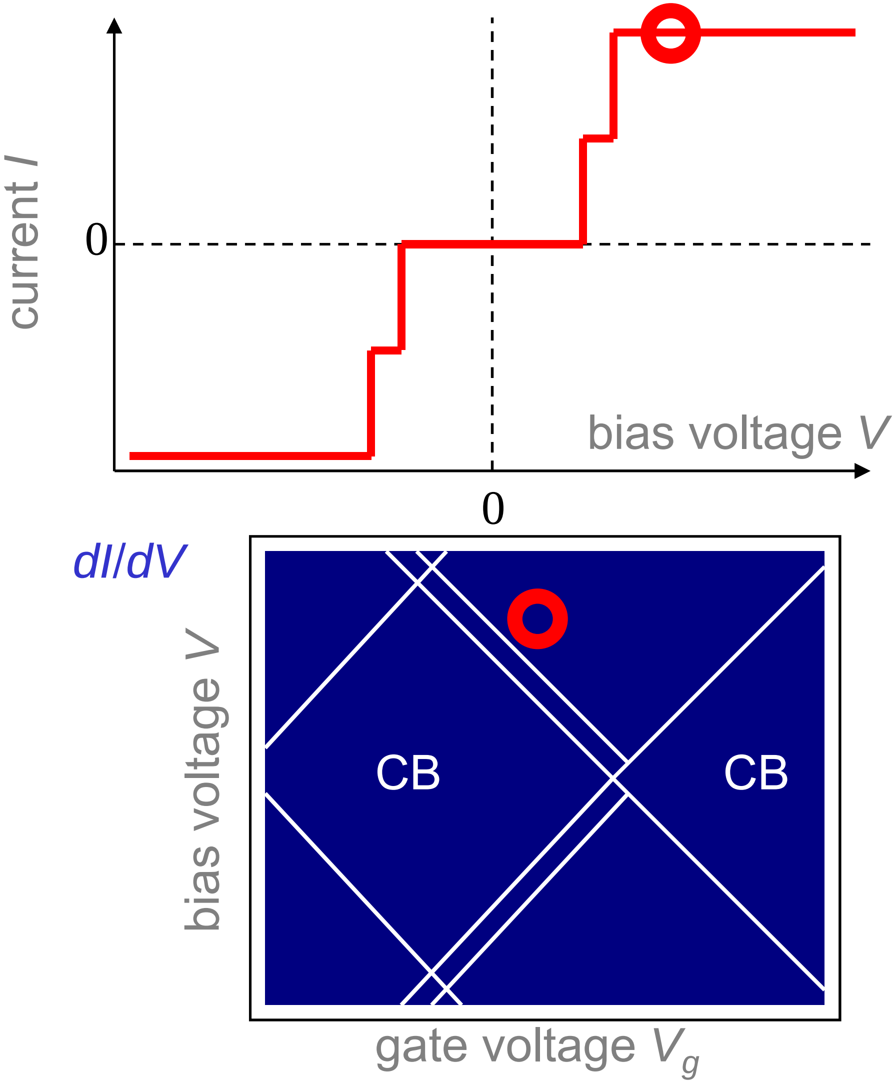


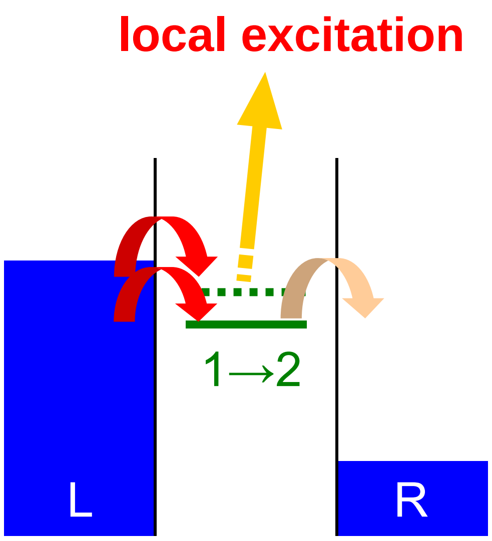




tunneling

Generic behavior described by rate equations





inelastic tunneling
(vibration, spin flip)

characteristic for
molecules

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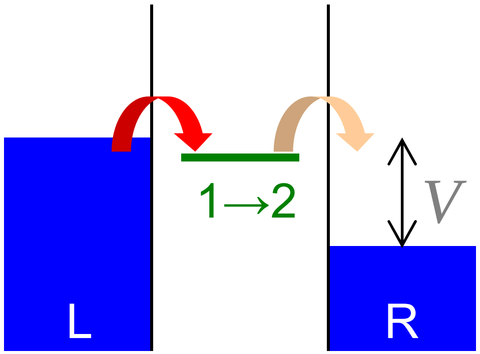
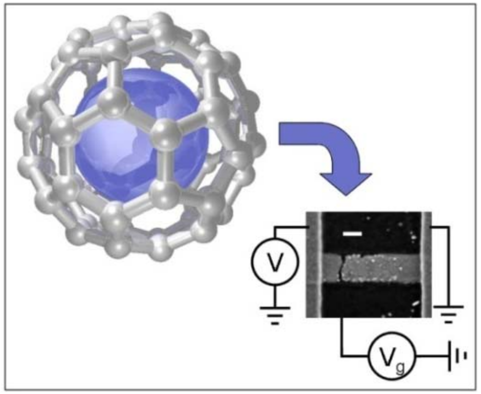


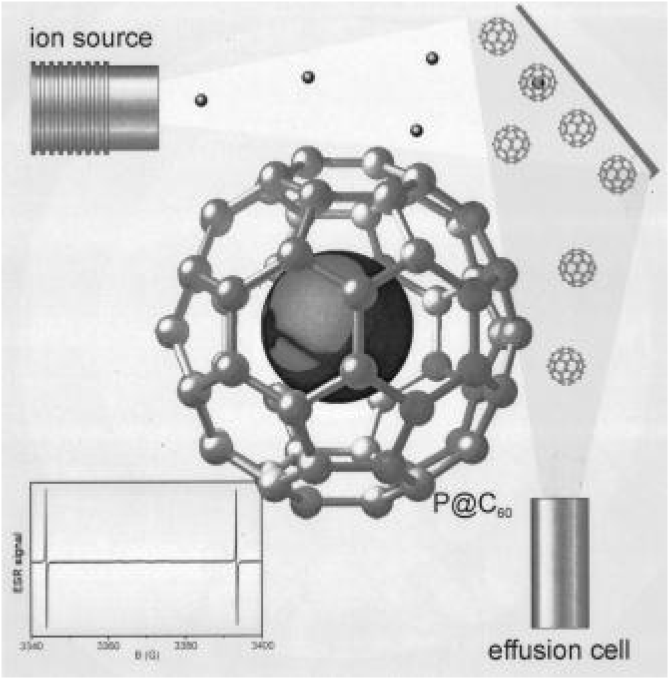


Endohedral $N@C_{60}$

- nitrogen atom located at center of C_{60}
- nitrogen retains spin $S_N = 3/2$ (Hund's 1st rule)

production by Harneit group (FU Berlin) using

- ion implantation
- enrichment / mass separation



Larsson *et al.*, J. Chem. Phys. **116**, 7849 (2002)
(shown for phosphorus)

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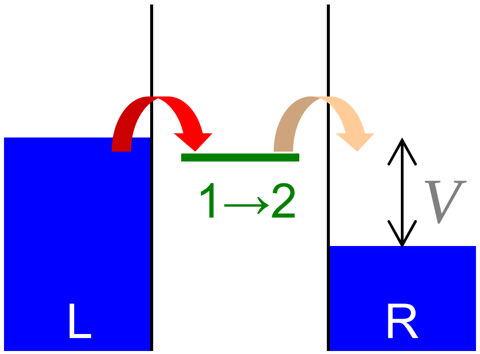

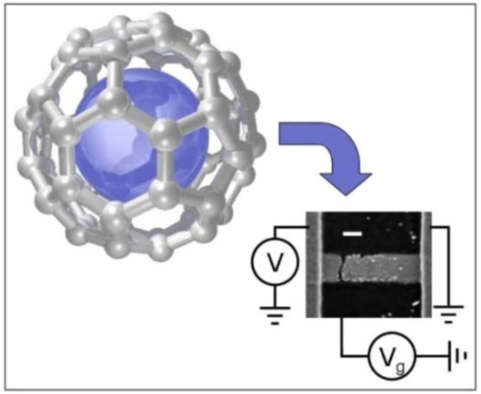

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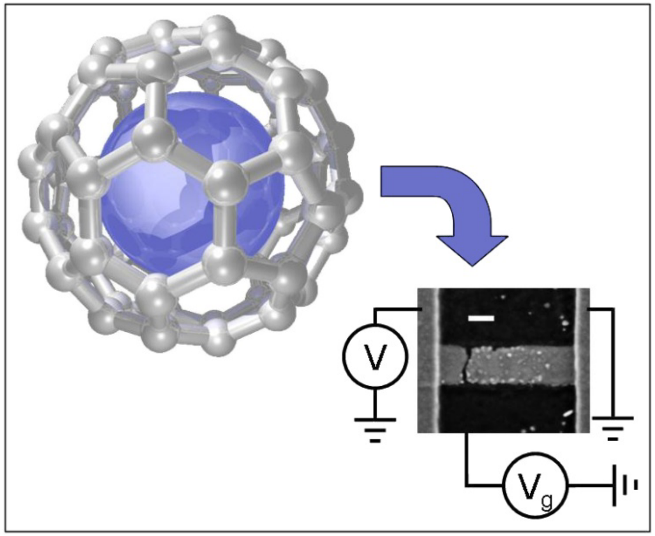
Motivation: Hope to observe inelastic tunneling due to coupling to molecular spin

earlier calculations by F. Elste and C.T., PRB **71**, 155403 (2005)



$N@C_{60}$ in Pt break junctions
(Ralph group, Cornell university)

J. E. Grose, E. Tam, C.T., M. Scheloske, B. Ulgut, J. J. Parks, H. D. Abruña, W. Harneit, and D. C. Ralph, Nature Materials **7**, 884 (2008)

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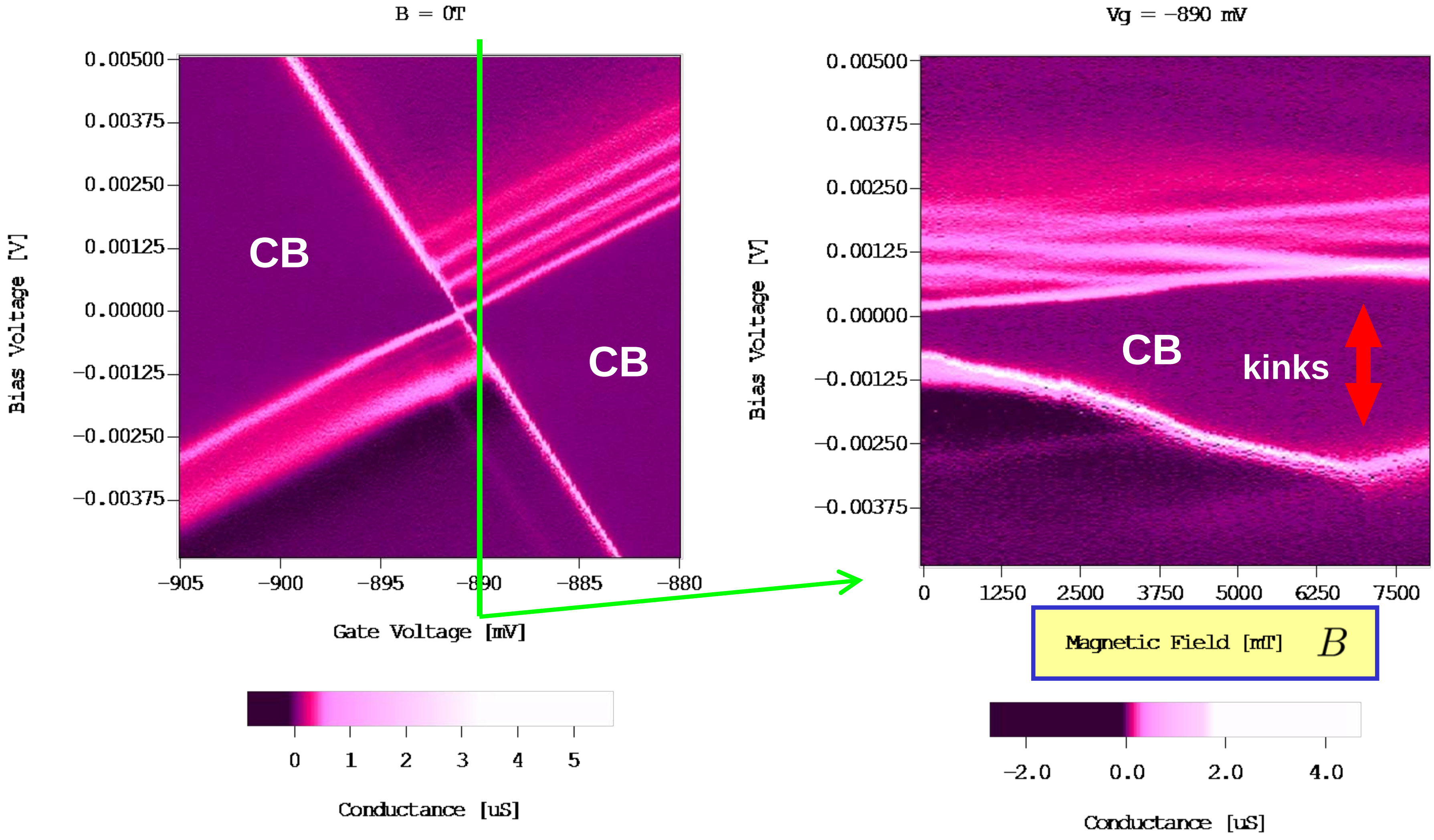
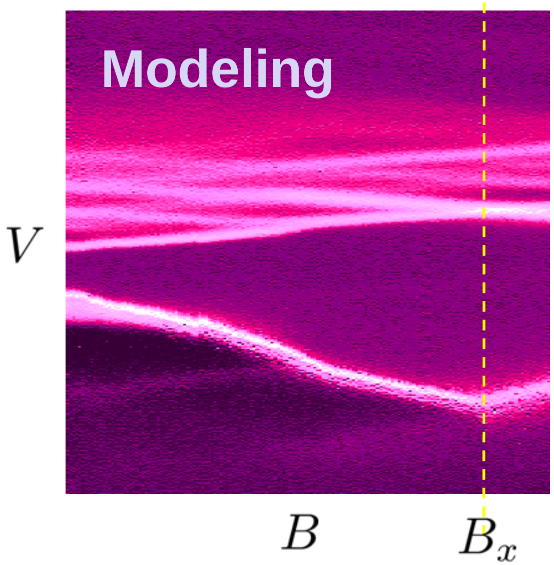
Differential conductance: experiment



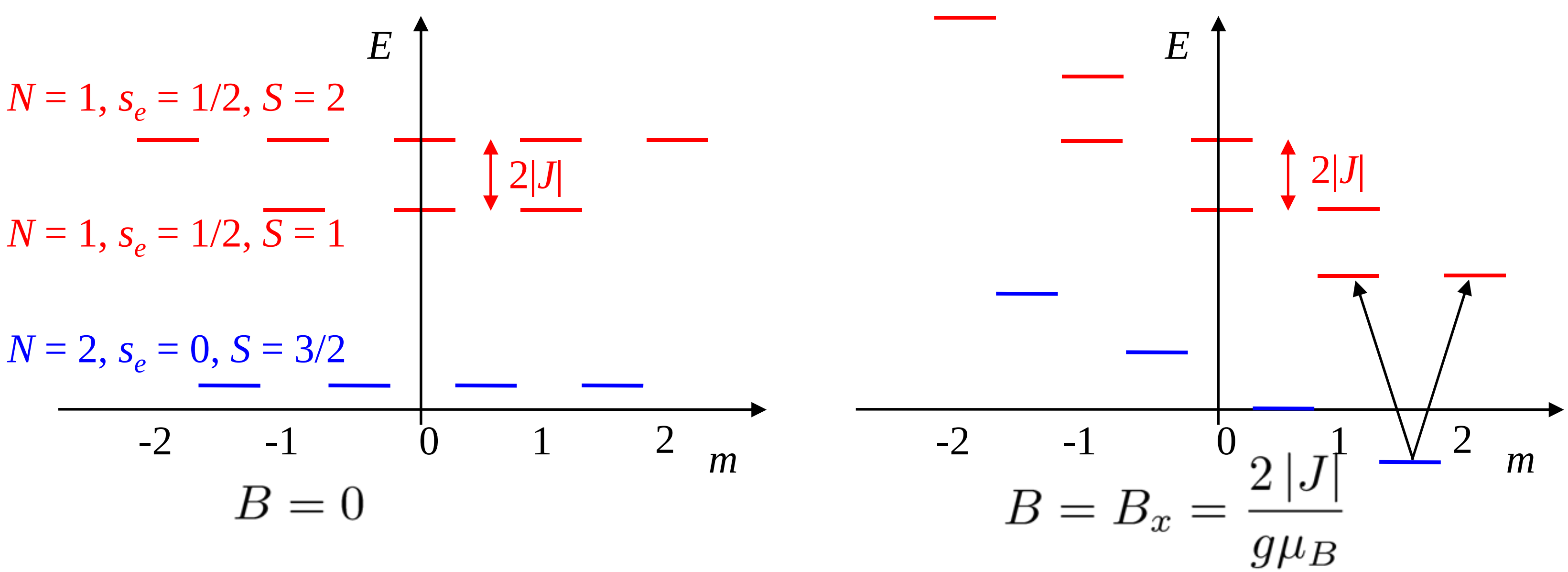


$$H_{\mathrm{dot,\,el}} = (\epsilon - eV_g^*)\sum_\sigma a_\sigma^\dagger a_\sigma + U\, a_\uparrow^\dagger a_\uparrow a_\downarrow^\dagger a_\downarrow$$
$$- J\,\mathbf{s}_e \cdot \mathbf{S}_N - g\mu_B B\,(s_e^z + S_N^z)$$

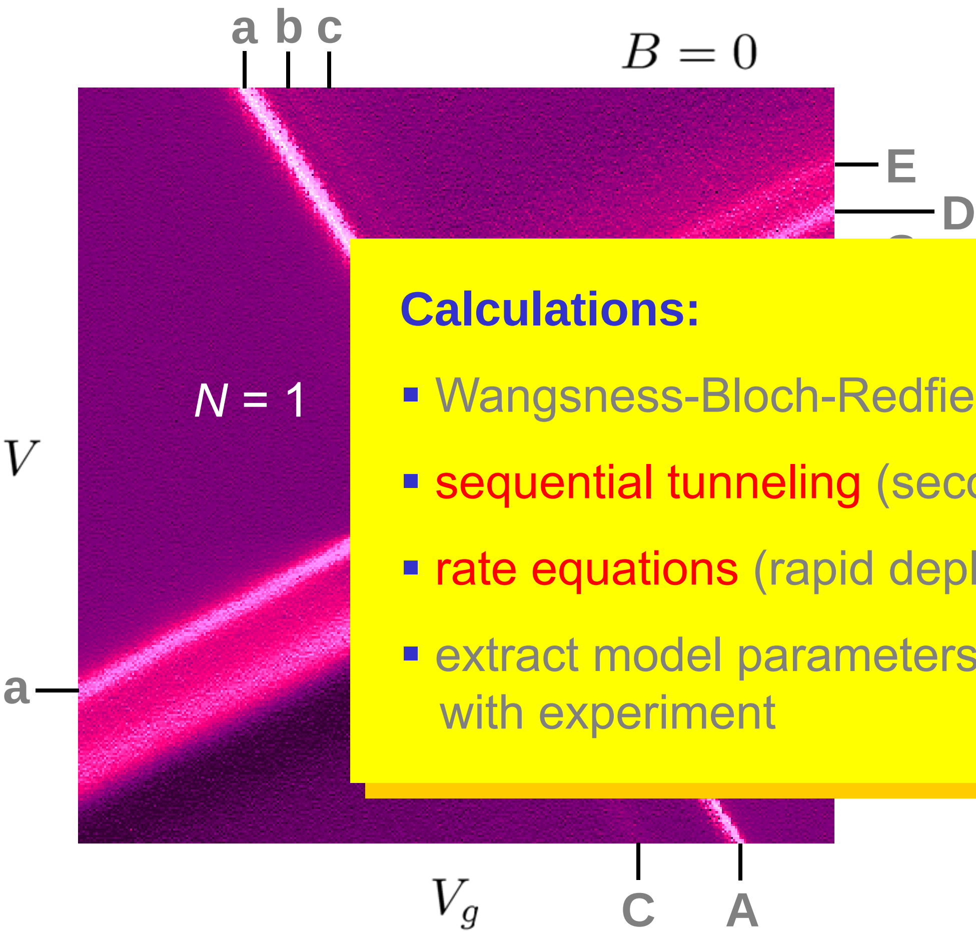
$V_g^* = \alpha V_g + \beta_L V$: local potential (asym. coupling)

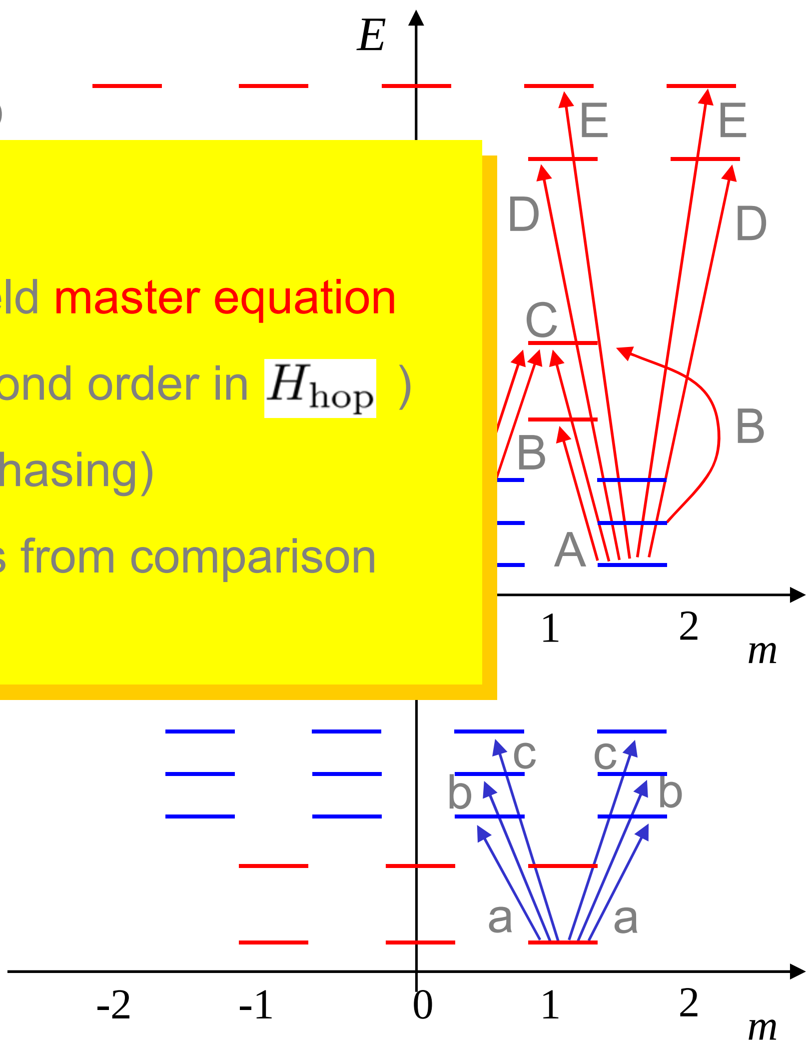
U : Coulomb repulsion on C_{60}

$J < 0$: exchange between electron and N spin



Fine structure: very soft oscillations?





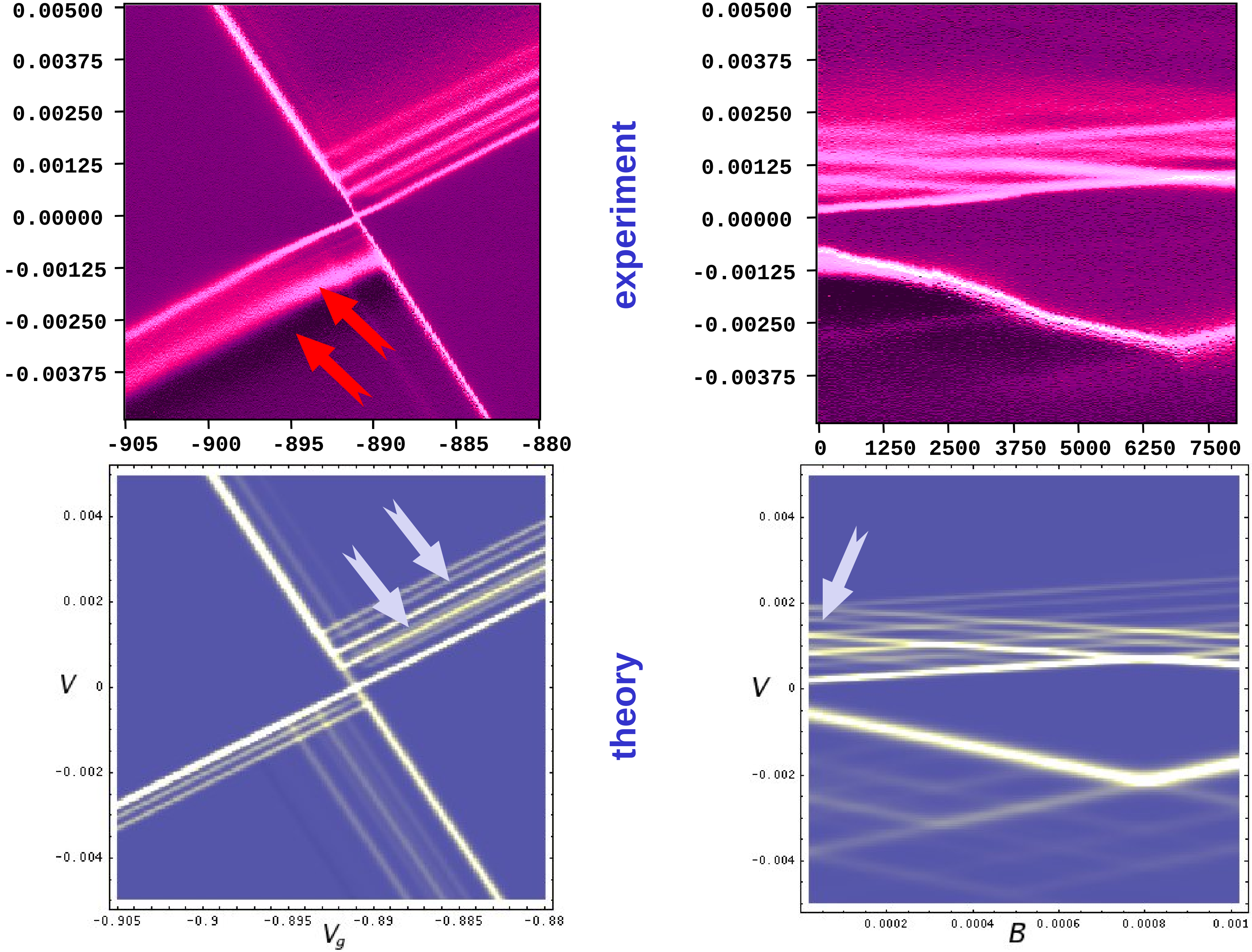
Calculations:

- Wangsness-Bloch-Redfield master equation
- sequential tunneling (second order in H_{hop})
- rate equations (rapid dephasing)
- extract model parameters from comparison with experiment

model makes predictions such as

$$\Delta E_A + \Delta E_b + \Delta E_B = \Delta E$$

OK


experiment
theory
0.00500
0.00375
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0.00125
0.00000
-0.00125
-0.00250
-0.00375
-905
-900
-895
-890
-885
-880
0
1250
2500
3750
5000
6250
7500
V
0.004
0.002
0
-0.002
-0.004
-0.905
-0.9
-0.895
-0.89
-0.885
-0.88
V_g
B
0.0002
0.0004
0.0006
0.0008
0.001

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Acknowledgements

F. Elste	McGill U
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