Magnetic N@C\textsubscript{60} single-molecule transistors

Towards modeling of real devices

Carsten Timm

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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

\[ \overline{A}_{\text{dot}}(t) = \overline{I}(t) = ? \]

- dot observable
- current

\[ \overline{A}_{\text{dot}}(t) = \text{Tr} \, \rho(t) \, A_{\text{dot}} \]

with the density operator

\[ \rho(t) \cong \rho_{\text{dot}}(t) \otimes \rho_{\text{leads}}^0 \]

Cannot solve this because \( H \) is complicated!

Now what?
$A_{\text{dot}}$ only depends on the dot: 

$$A_{\text{dot}}(t) = \text{Tr} \, \rho_{\text{dot}}(t) \, A_{\text{dot}}$$

with reduced density operator (in “small” dot Hilbert space)

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

basis of lead (reservoir) states only

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

**The Master Equation!**

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

$$\frac{d\rho}{dt} = -i \, [H, \rho] \quad \implies \quad \frac{d}{dt} \rho_{\text{dot}} = -i \, \text{tr}_{\text{leads}}[H, \rho(t)]$$
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 \( H_{\text{hop}} \)
- assume product state with leads in equilibrium at time \( t \): \( \rho(t) \cong \rho_{\text{dot}}(t) \otimes \rho_{\text{leads}}^0 \)

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

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

Wangsness-Bloch-Redfield master equation

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

→ time evolution not unitary, includes relaxation

see C.T., PRB 77, 195416 (2008)
Case 1: single reservoir (particle & energy bath)
dot approaches equilibrium for $t \to \infty$:

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

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:  \( H_{\text{dot}} |m\rangle = E_m |m\rangle \)

*If* off-diagonal components of \( \rho_{\text{dot}} \) in basis \( \{ |m\rangle \} \) relax rapidly (rapid dephasing):

sufficient to keep only diagonal components

\[
P_m \equiv (m | \rho_{\text{dot}} | m) \quad \text{probabilities of dot states } |m\rangle
\]

obtain rate equations

\[
\frac{dP_m}{dt} = \sum_n \left( R_{n \rightarrow m} P_n - R_{m \rightarrow n} P_m \right)_{\text{in out}}
\]

\[ P_m(t) \quad \text{observables, e.g. } I(t) \equiv \overline{I}(t) \]
**Generic behavior** described by rate equations

- $dI/dV$
- $V$
- $I$
- $V_g$
- $V$
- $CB$
- L
- R

very small current: Coulomb blockade
Generic behavior described by rate equations

dI/dV

bias voltage V

current /

V

gate voltage V_g

CB

1→2

tunneling
Generic behavior described by rate equations

Inelastic tunneling (vibration, spin flip)
characteristic for molecules
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Endohedral $\text{N}@\text{C}_{60}$

- nitrogen atom located at center of $\text{C}_{60}$
- nitrogen retains spin $S_N = 3/2$ (Hund’s 1st rule)

production by Harneit group (FU Berlin) using
- ion implantation
- enrichment / mass separation

(shown for phosphorus)
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**Motivation:** Hope to observe inelastic tunneling due to coupling to molecular spin


Differential conductance: experiment
Modeling

\[ H_{\text{dot, el}} = (\epsilon - eV^*_g) \sum_{\sigma} a_{\sigma}^\dagger a_{\sigma} + U a_{\uparrow}^\dagger a_{\uparrow} a_{\downarrow}^\dagger a_{\downarrow} \]
\[ -J s_e \cdot S_N - g\mu_B B (s_e^z + S_N^z) \]
\[ V_g^* = \alpha V_g + \beta g_L V : \text{local potential (asym. coupling)} \]
\[ U : \text{Coulomb repulsion on } C_{60} \]
\[ J < 0 : \text{exchange between electron and } N \text{ spin} \]

\[ N = 1, s_e = 1/2, S = 2 \]
\[ N = 1, s_e = 1/2, S = 1 \]
\[ N = 2, s_e = 0, S = 3/2 \]

\[ B = 0 \]
\[ B = B_x = \frac{2|J|}{g\mu_B} \]
Calculations:
- Wangsness-Bloch-Redfield master equation
- sequential tunneling (second order in $H_{\text{hop}}$)
- 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
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