Forschungszentrum Karlsruhe
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Tunneling transport through molecules and coupled quantum dots

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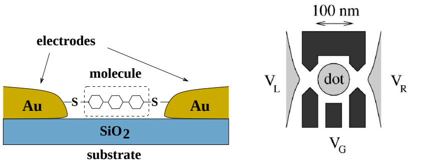
I. Motivation

- Quantum dots and molecules as potential building blocks for future nanoelectronic devices
- Problem: coupling of molecule to electrodes unknown and not easily controllable in nanoscopic devices.
- Combined measurement of current and shot noise provides additional information about couplings, interaction, etc.

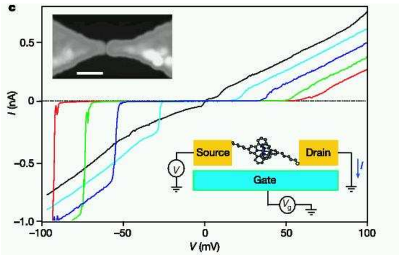
II. Physical Realization



l.h.s.: deposition of molecule between two fixed electrodes (Reed et al., Science **278**, 252 (1997))

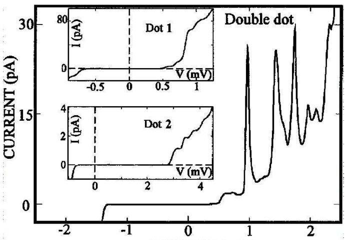
r.h.s.: schematic picture of a lateral quantum dot structure

III. Experiments



J. Park , A.N. Pasupathy, *et.al.*, Nature **417**, 722 (2002)

- electrodes probed via electron beam lithography and electromigration
- molecule is a Co ion bonded to ligands → coupling to the electrodes can be controlled

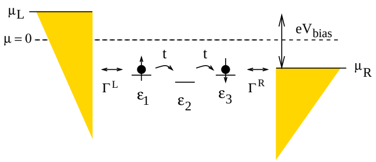


van der Wiel *et al.*, Rev. Mod. Phys. **75**, 1 (2003)

- lateral coupled quantum dots (”artificial molecules”)
- single-electron phenomena such as Coulomb blockade observed

IV. Model

Sketch of couplings for three coupled quantum dots



- $\hat{H} = \hat{H}_{\mathrm{L}} + \hat{H}_{\mathrm{R}} + \hat{H}_{\mathrm{dots}} + \hat{H}_{\mathrm{T,L}} + \hat{H}_{\mathrm{T,R}}$ with:

$$\hat{H}_r = \sum_{k\sigma} \varepsilon_{k\sigma r} a^\dagger_{k\sigma r} a_{k\sigma r},$$
$$\hat{H}_{\mathrm{T},r} = \sum_{ik\sigma} (t^r_i a^\dagger_{k\sigma r} c_{i\sigma} + h.c.),$$
$$\hat{H}_{\mathrm{dots}} = \sum_{i\sigma} \varepsilon_i n_{i\sigma} - t \sum_{<ij>\sigma} (c^\dagger_{i\sigma} c_{j\sigma} + h.c.)$$
$$+ U \sum_i n_{i\uparrow} n_{i\downarrow} + U_{nn} \sum_{<ij>\sigma\sigma'} n_{i\sigma} n_{j\sigma'}$$

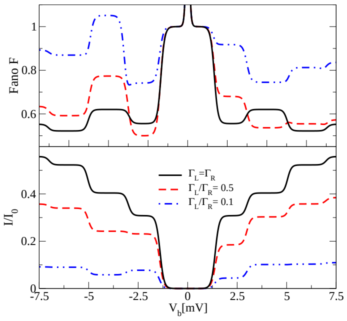
- $\hat{H}_{\mathrm{L}}$, $\hat{H}_{\mathrm{R}}$: left and right electrode with non-interacting electrons ($r = \mathrm{L}, \mathrm{R}$).
- $\hat{H}_{\mathrm{dots}}$: molecule or dot term with i (spin-dependent) molecular sites ($n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$).
- $\hat{H}_{\mathrm{T,L}}$, $\hat{H}_{\mathrm{T,R}}$: tunneling between the dot system and electrodes.
- Energies: level energies (ε_i), intra-dot (U) and inter-dot (U_{nn}) Coulomb repulsion, kinetic (hopping) energy t)
- coupling strength $\Gamma^r_i = 2\Pi |t^r_i|^2 \rho_e$ with t^r_i being the tunneling amplitude and ρ_e is DOS

V. Numerical Results

A. Two coupled quantum dots

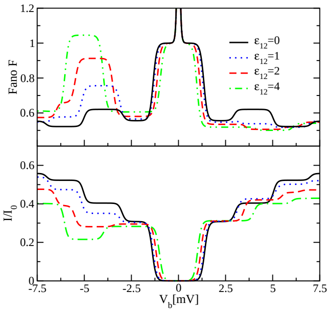
Fano factor $F = \frac{S}{2eI}$ for symmetric bias voltage and $\varepsilon_1 = \varepsilon_2 = -5.5, t = 2, U = 10, U_{nn} = 5, k_{\mathrm{B}}T = 0.05$ (all units in meV). Step positions determined by energy parameters and plateau heights by couplings $\Gamma_{\mathrm{L}}, \Gamma_{\mathrm{R}}$. The total coupling scale is set to $\Gamma_R = 0.05\, k_B T$.

Asymmetric coupling $\Gamma_{\mathrm{L}} \neq \Gamma_{\mathrm{R}}$



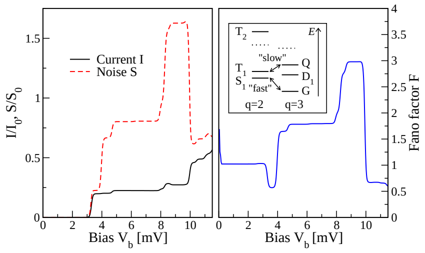
For strong asymmetries super-Poissonian Noise (F>1) and negative differential conductance (NDC) develop at the same bias (energy) positions

Strong level detuning, in particular $\varepsilon_{12} = \varepsilon_1 - \varepsilon_2 > t$, leads to super-Poissonian noise and NDC



B. Three coupled quantum dots

Strong non-local Coulomb interactions $U_{nn} > t$
Current and Noise for symmetric coupling $\Gamma_{\mathrm{R}} = \Gamma_L$ with parameters $\varepsilon_i = -10$, $t = 2$, $U = 10$, $U_{nn} = 5$, $k_{\mathrm{B}}T = 0.025$ (units in meV) and more complex many body wavefunctions (Singlets S, Doublets D, Triplets T, Quadruplets Q)

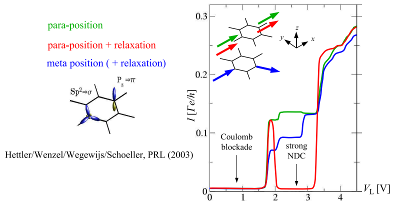


Super-Poissonian noise and Fano factors without simultanous suppression of the current

→ competition of “fast” and “slow” transport channels leads to strongly enhanced noise
→ effect depends on the ratio $\frac{U_{nn}}{t}$ and is entirely due to internal electronic level structure

C. Towards a description of Molecules

Six site model for aromatic molecules like benzene rings including relaxation processes of photons



Hettler/Wenzel/Wegewijs/Schoeller, PRL (2003)

Transport highly sensitive to different coupling positions (para and meta position)

In the case of paraposition strong NDC is observed → occupation of a blocking state of the molecule

References:
Thielmann *et. al.*, Phys. Rev. Lett. **95**, 146806 (2005), Aghassi *et. al.*, Phys. Rev. B **73**, 195323 (2006), Aghassi *et. al.* cond-mat/0505345 (to be published in Appl. Phys. Lett.)