Transport through a molecular device: molecular symmetry, Coulomb interaction and relaxation processes

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We predict a *negative differential conductance* (NDC) effect in the nonlinear transport through a molecular device. The device consists of a benzene ring which is *weakly* coupled to electrodes (via intermediate molecules which "cut" the π conjugation between the ring and the electrodes [1]) at the para-positions (1st and 4th carbon atom along the ring). The effect is based on

- the symmetry properties of the planar molecule with respect to the transport direction (mirror reflection symmetry w.r.t. plane perpendicular to molecule and containing the transport axis)
- the strong e-e interactions on the molecule (including Coulomb blockade effects)
- the relaxation of molecular and anionic excited states due the coupling of the electrons to photons (spontanous decay)

The flow of current is blocked by the spontaneous decay of the *anion* from an *excited* state to a lower lying, stable state of the anion which does not couple to the electron tunneling, a "blocking state". The effect is generic: other molecules may have similar current-switching properties due to such blocking states of the charged molecule. An effective *interacting* Hamiltonian for the π-electrons (derived from an electronic structure calculation) serves as input for the transport calculation (perturbation theory in electron tunneling/Golden-Rule). The effect of an applied electric field on the molecular states (screening) is also investigated. We furthermore compare the results for the case where the molecule is coupled at the para-positions and at the meta-positions (1st and 3rd carbon atom along the ring). We discuss the robustness of the predicted effects and possible spoilers.

Finally, we report our progress in extending the approach to treat Kondo-type of effects in certain molecules which have recently been investigated experimentally [2].

[1] M. Mayor *et al*, Angew. Chem. **114**, 1228 (2002).
[2] J. Park, A. N. Pasupathy *et.al.*, Nature **417**, 722 (2002).