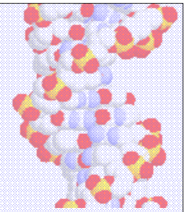
Workshop on computer simulations of soft matter and biosystems, Heidelberg, March 14-16 (2007)

Conduction properties of DNA wires: Hamiltonian model approaches

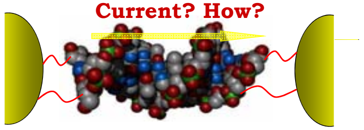
R. Gutierrez and G. Cuniberti, University of Regensburg



State-of-the-art

DNA: a molecular wire?

Current? How?



Controversial electrical *transport* experiments:
- insulator?
- semiconductor?
- metallic?

High sensitivity to
- metal-molecule contact
- base-pair sequence
- environmental effects
- charge injection mechanisms
- sample preparation
- single molecules vs. bundles

Selected MC-group publications

- **Semiconducting behavior**
  -G. Cuniberti, L. Craco, D. Porath, and C. Dekker
  Backbone-induced semiconducting behavior in short DNA wires, Phys. Rev. B **65**, 241314(R) (2002)

  -J. Yi, *Conduction of DNA molecules: A charge-ladder model,* Phys. Rev. B **68**, 193103 (2003)

- **Coupling to vibrons and to the environment**
  -R. Gutierrez, S. Mandal and G. Cuniberti
  Quantum transport through a DNA wire in a dissipative environment, Nano Lett. **5**, 1093 (2005)

  -R. Gutierrez, S. Mandal and G. Cuniberti
  Dissipative Effects in the Electronic Transport through DNA molecular wires, Phys. Rev. B **71**, 235116 (2005)

  -R. Gutierrez, S. Mohapatra, H. Cohen, D. Porath, and G. Cuniberti
  Inelastic quantum transport in a ladder model: application to DNA conduction, Phys. Rev. B **74**, 235105 (2006)

- **STS-spectroscopy**
  -Y. Calev, H. Cohen, G. Cuniberti, A. Nitzan and D. Porath
  Tight-binding description of the STM image of molecular chains, Israel Journal of Chemistry 44, 133 (2004)

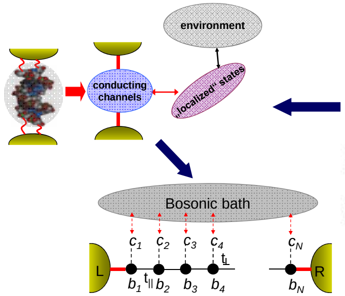
  -E. Shapir, J. Yi, H. Cohen, A. Kotlyar, G. Cuniberti and D. Porath
  The puzzle of contrast inversion in DNA STM imaging, Journal of Phys. Chem. B **109**, 14270 (2005)

  -D. Ryndyk and G. Cuniberti, in preparation (2007)

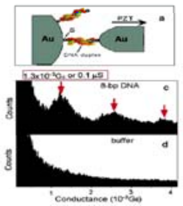
- **Reviews**
  -D. Porath, G. Cuniberti, and R. Di Felice
  Charge transport in DNA-based devices, Topics in Current Chemistry **237**, 183 (2004).
  -R. Gutierrez, D, Porath, and G. Cuniberti
  DNA conduction: the issue of static disorder, dynamic fluctuations and environmental effects, Charge Transport in Disordered Solids with Applications in Electronics, edited by S. Baranovski, Wiley-CH (2006)

DNA conduction in a dissipative environment

R. Gutierrez, S. Mandal, and G. Cuniberti, Nano Lett. **5**, 1093 (2005); ibidem., Phys. Rev. B **71**, 235116 (2005).



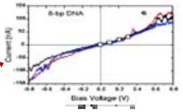
Motivation

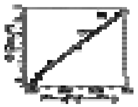


single molecules probed

ohmic-like I-V curves

power-law conductance scaling with DNA length





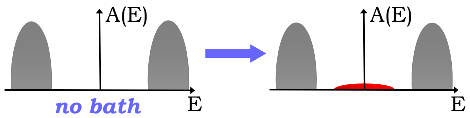
Direct Conductance Measurement of Single DNA Molecules in Aqueous Solution
B. Xu, P. Zhang, X. Li, N. Tao, Nano Lett. **4**, 1105 (2004).

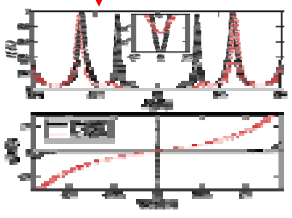
Model Hamiltonian

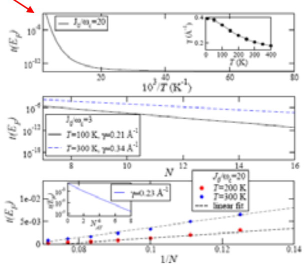
$$\mathcal{H} = \epsilon_b \sum_j b_j^\dagger b_j - t_{||} \sum_j \left(b_j^\dagger b_{j+1} + \mathrm{H.c.}\right) + \epsilon \sum_j c_j^\dagger c_j - t_\perp \sum_j \left(b_j^\dagger c_j + \mathrm{H.c.}\right) + \sum_{\mathbf{k}\in \mathrm{L,R},\sigma} \epsilon_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma}^\dagger d_{\mathbf{k}\sigma} + \sum_{\mathbf{k}\in \mathrm{L},\sigma} \left(V_{\mathbf{k},1} d_{\mathbf{k}\sigma}^\dagger b_1 + \mathrm{H.c.}\right) + \sum_{\mathbf{k}\in \mathrm{R},\sigma} \left(V_{\mathbf{k},N} d_{\mathbf{k}\sigma}^\dagger b_N + \mathrm{H.c.}\right) + \sum_\alpha \Omega_\alpha B_\alpha^\dagger B_\alpha + \sum_{\alpha,j} \lambda_\alpha c_j^\dagger c_j \left(B_\alpha + B_\alpha^\dagger\right).$$

Results

Qualitative behavior of the wire spectral density at ***low energies***
Selfenergy leads to bath-induced (*virtual*) polaronic states *inside* the electronic band gap, which support *activated transport* at low injection energies.



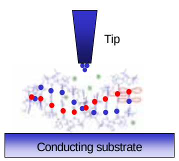




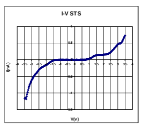
- *Arrhenius-like* behavior of the linear conductance *g*
- *Weak* exponential length scaling $g \sim e^{-\gamma N}$ going over to
- *Power-law scaling* $g \sim N^{-\gamma}$, $\gamma \sim 0.1\text{-}0.2\ Å^{-1}$ with *increasing* system-bath coupling

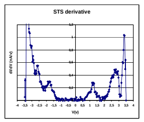
STS-Spectroscopy of Poly(GC) wires

D. Ryndyk and G. Cuniberti, submitted (2006)

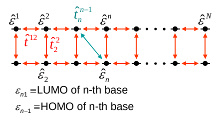


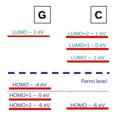
General features
- single poly(GC) wires
- weak coupling to the substrate and the STM tip
- two tunnel junctions
- tunnel currents (~1 nA at 3V)
- gap ~ 2.5 V
- asymmetry and steps/peaks in I-V curves

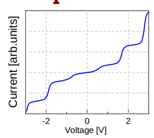


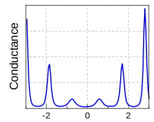


Model and comparison to experiments





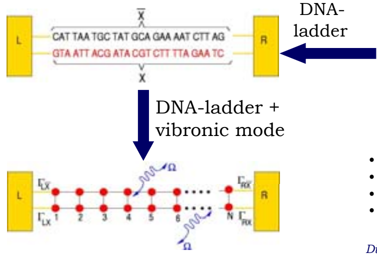


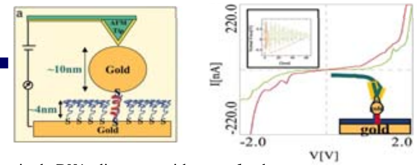


Inelastic transport in DNA wires

R. Gutierrez, S. Mohapatra, D. Porath, and G. Cuniberti, Phys. Rev. **B** 74, 235105 (2006)

Motivation





- single DNA oligomers with ***complex*** base sequence
- ***strong*** coupling to the contacts via SH-groups
- ***nonlinear*** I-Vs, very high currents (~220 nA at 2 V)
- purely coherent band-like transport seems unlikely in view of the random sequence

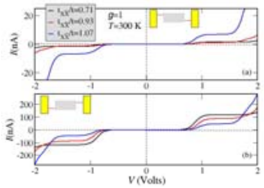
Direct measurement of electrical transport through single DNA molecules of complex sequence, H. Cohen, C. Nogues, R. Naaman, D. Porath, Proc. Natl. Acad. Sci. USA **102**, 11589 (2005).

Model Hamiltonian

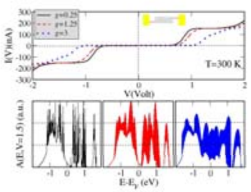
$$\mathcal{H} = \sum_{r=X,Y}\sum_\ell \epsilon_{r,\ell} b_{r,\ell}^\dagger b_{r,\ell} - \sum_{r=X,Y}\sum_\ell t_{r,\ell,\ell+1}[b_{r,\ell}^\dagger b_{r,\ell+1} + h.c.] - \sum_\ell t_{\perp,\ell}[b_{X,\ell}^\dagger b_{Y,\ell} + h.c.] + \sum_{k\in L}[t_{k,X} c_k^\dagger b_{X,1} + h.c.] + \sum_{k\in L}[t_{k,Y} c_k^\dagger b_{Y,1} + h.c.] + \sum_{k\in R}[t_{k,X} c_k^\dagger b_{X,N} + h.c.] + \sum_{k\in R}[t_{k,Y} c_k^\dagger b_{Y,N} + h.c.] + \sum_\alpha \Omega_\alpha B_\alpha^\dagger B_\alpha + \sum_{\alpha,r,\ell} \lambda_\alpha b_{r,\ell}^\dagger b_{r,\ell}(B_\alpha + B_\alpha^\dagger)$$

Results

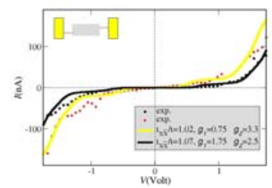
Dramatic sensitivity to
-electrodes-DNA contact symmetry
-inter-strand coupling t_{XY}



-Phonon satellites + zero-phonon lines strongly overlapping
-Phonon blockade at low bias



-Two-phonon model: semi-quantitative agreement with experiments on suspended DNA



Outlook
- Effective electronic Hamiltonian in **noisy environments** and relation to molecular dynamics simulations
- Role of correlations in metallic-DNA: extended Hubbard models
- Non-equilibrium **incoherent** transport via Keldysh Green functions
- Inelastic STS of DNA oligomers on substrates

Goal: Realistic description of DNA quantum transport via a multiscale modelling approach

Systems
- Poly(GC), Poly(AT) and G4-quartets
- Non-periodic and random (biological) sequences
- M-doped Poly(GC), Poly(AT) and G4 with M=Cu,Zn,Ag

Methods
- DFT and molecular dynamics (MD)
- Keldysh Green function techniques
- reduced density matrix theory

Research partners

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Rosa Di Felice (University of Modena, Italy)
Marcus Elstner (TU Braunschweig)
Joshua Jortner (Tel Aviv University, Israel)
Danny Porath (Hebrew University, Israel)
Julio Gomez (Universidad Autonoma, Spain)
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