Quantum transport in DNA wires: Influence of a dissipative environment

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Outline

- Why DNA?
- Electronic transport in DNA: a bird’s eye view
- DNA wire in water: a Hamiltonian model
- Formalism and approximations
- Strong dissipative limit
- Conclusions
Why DNA?

- **Groundbreaking**: repair of oxidative damage
- ET over long distances (∼40 Å)
  
  (C. J. Murphy et al., Science (1993))

- Molecular electronics ⇒ potential applications
  as template (self-recognition and assembling)
  as molecular wire (M-DNA, poly(GC))
Electronic transport in DNA: a bird’s eye view

- **Experiments**: DNA is insulator, metal, semiconductor
  - sample preparation and experimental conditions are crucial
    - (dry vs. aqueous environment, metal-molecule contacts, single molecules vs. bundles ...)

- **Theory**: Variety of factors modifying charge propagation:
  - static disorder, dynamical disorder, environment (hydration shell, counterions)

see: D. Porath, G. Cuniberti, and R. Di Felice,

*Charge Transport in DNA-Based Devices*

Transport in single Poly(GC) oligomers in water


metallic behaviour!

large currents!
Transport in single Poly(GC) oligomers in water

\[ g_{GC} \sim \frac{1}{L} \]

\[ g_{GC-AT} \sim e^{-\gamma L} \]
\[ \gamma \sim 0.43 \text{ Å}^{-1} \]

Ab initio (H. Wang et al. PRL (2004)): dry Poly(GC) \( \sim e^{-\gamma L}, \gamma \sim 1.5 \text{ Å}^{-1} \)

Algebraic behaviour induced by the environment !?
DNA wire in water: a Hamiltonian model

- *ab initio* calculations (e.g. E. Artacho *et al.*, Mol. Phys. (2003))

  (i) decoupled HOMO/LUMO channels

  (ii) backbones non conducting

\[
\mathcal{H} = \sum_j \epsilon_j b_j^\dagger b_j - \frac{t_{\parallel}}{\langle i,j \rangle} \sum_{i,j} \left( b_i^\dagger b_j + \text{H.c.} \right) \\
+ \sum_j \epsilon_j d_j^\dagger d_j - t_{\perp} \sum_j \left( b_j^\dagger d_j + \text{H.c.} \right) \\
+ \sum_\alpha \Omega_\alpha B_\alpha^\dagger B_\alpha + \sum_{\alpha,j} \lambda_\alpha d_j^\dagger d_j \left( B_\alpha + B_\alpha^\dagger \right) \\
+ \mathcal{H}_{\text{leads}}
\]
Formalism and approximations

- Green function techniques
- Phonon bath $\sim$ continuous frequency spectrum $J(\omega) \sim \left(\frac{\omega}{\omega_c}\right)^S$

- neglect nonequilibrium effects ($eV \rightarrow 0$)
- no inelastic electron tunneling, $E_{in}^{el} = E_{out}^{el}$

$\sim$ Landauer-like expression:

$$t(E) = Tr\left[G_W^\dagger(E)\Gamma_R G_W(E) \Gamma_L\right]$$

$t(E)$ includes full interaction with the bath $\sim$ decoherence
Low-bias, strong dissipative limit

New $k_B T$-dependent electronic manifold around $E_F = 0$

Bath-selfenergy $\Sigma_B(E)$:

$\text{Re } \Sigma_B(E) \sim$ renormalization and/or new states (polaron band)

$\text{Im } \Sigma_B(E)$ ("friction") $\sim$ decoherence, strong suppression of the polaron band

$\sim$ incoherent polaronic band $\sim$ pseudo-gap opening
Transmission $t(E)$ and low-bias current $I(V)$

- **pseudo-gap** increases with temperature
- **incoherent DOS** also increases with temperature
- $I(V)$ displays “metallic” behaviour at high $k_B T$
Temperature dependence of $t(E_F)$ (Arrhenius plot)

Activated behaviour

$\sim$ phonon assisted hopping
Scaling of $t(E_F)$ with the chain length $L = Na_{bp}$

- Increasing coupling to the bath
  
  \[ t_F \sim e^{-\gamma L} \rightarrow \text{algebraic } t_F \sim L^{-\alpha} \text{ dependence} \]

- Exponential dependence is related to virtual tunneling through pseudo-gap ($\gamma \ll 1 \text{ Å}^{-1}$)

- Introduction of a barrier $\sim (AT)_n$ pairs, enforces exp-dependence
Structural base-pair fluctuations

Random on-site energies drawn from Gaussian distribution $P(\epsilon) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\epsilon^2/2\sigma^2}$

“Thermal” disorder does not appreciably affect the pseudo-gap formation
Conclusions

- Environment drastically affects charge transport
- **Strong coupling** regime:
  - (i) bath-induced pseudo-gap
  - (ii) finite $k_B T$-dependent DOS near $E_F$
  - $\sim$ activated behaviour
  - $\sim$ weak exponential or algebraic $L$-dep.
- Contact to Xue *et al.* (2004) experiments
  - (i) (too ?) large currents $\sim 50 - 200 \, nA$
  - (ii) (bath-induced) algebraic $L$-dependence

Perspectives

- parameter estimation
- Sequence dependence
  - (Klotsa *et al.* ICPS27 (2004), Roche *et al.* PRL (2003))
- Nondiagonal EPI