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-based molecular wires in water
  (i) Motivation: Poly(GC) oligomers in aqueous solution
  (ii) The model Hamiltonian
  (iii) Green functions, approximations, current etc
  (iv) Results: low-bias, strong coupling limit (wet DNA)

• Conclusions and Outlook
Why DNA?

- Groundbreaking: repair of oxidative damage
  \[ \leadsto \text{ET over long distances (\sim 40 \text{ Å})} \]
  \[ \text{(C. J. Murphy et al., Science (1993))} \]

- Molecular electronics \Rightarrow potential applications
  as template (self-recognition and assembling)
  as molecular wire (M-DNA, poly(GC))

M. Hazani et al., CPL (2004)
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

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

Charge Transport in DNA-Based Devices
Motivation: Transport in single Poly(GC) oligomers in water (I)


“metallic” behaviour, large currents
Motivation: Transport in single Poly(GC) oligomers in water (II)

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

$\Rightarrow$ Algebraic behavior induced by the environment?
A minimal model for a DNA wire in “water”


\[
\mathcal{H} = \sum_j \epsilon_{b,j} b_j^\dagger b_j - t_\pi \sum_j \left( b_j^\dagger b_{j+1} + \text{H.c.} \right) + \sum_j \epsilon_j c_j^\dagger c_j - t_\perp \sum_j \left( b_j^\dagger c_j + \text{H.c.} \right) + \sum_\alpha \Omega_\alpha B_\alpha^\dagger B_\alpha + \sum_{\alpha,j} \lambda_\alpha c_j^\dagger c_j \left( B_\alpha + B_\alpha^\dagger \right) + \mathcal{H}_{\text{leads}} + \mathcal{H}_{\text{leads}}
\]
Green function techniques

- Polaron transformation: $\mathcal{H} \Rightarrow e^S \mathcal{H} e^{-S}$, $S = \sum_{\alpha,j} \frac{\lambda_\alpha}{\Omega_\alpha} c_j^\dagger c_j (B_\alpha - B_\alpha^\dagger)$

$$\rightarrow \sum_j (\epsilon_j + \sum_{\alpha} \frac{\lambda_\alpha^2}{\Omega_\alpha}) c_j^\dagger c_j \quad \rightarrow -t_\perp \sum_j [b_j^\dagger c_j \exp (-\sum_{\alpha} \frac{\lambda_\alpha}{\Omega_\alpha} (B_\alpha^\dagger - B_\alpha))] + \text{H. c.}$$

- Green functions ($\hbar = 1$)

$$G_{jl}(t) = -i \theta(t) \langle \{ b_j(t), b_{\ell}(0) \} \rangle$$

$$G^{-1}(E) = \frac{E 1 - \mathcal{H}_{\pi-\pi} - \sum_{L}(E) - \sum_{R}(E) - \frac{t_\perp^2 P(E)}{\Delta}}{\Delta}$$

$P_{\ell j}(E) = -i \delta_{\ell j} \theta(t) \langle \{ c_j(t) \mathcal{X}(t), c_{\ell}(0) \mathcal{X}(0) \} \rangle$

- Continuous bath frequency distribution ($N \to \infty$) $\sim$ spectral density:

$$J(\omega) = \sum_{\alpha} \lambda_\alpha^2 \delta(\omega - \Omega_\alpha) = \frac{J_0}{\omega_c} \omega \left( \frac{\omega}{\omega_c} \right)^{s-1} e^{-\omega/\omega_c} \theta(\omega)$$
The electric current

\[ I_L = -I_R \]

\[ I_L = \frac{2e}{\hbar} \int dE \left( \Sigma_L^\geq G^> - \Sigma_L^\leq G^< \right) \]

\[ I_L^{\text{el}} = \frac{2e}{\hbar} \int dE (f_L - f_R) \underbrace{\text{Tr}[G^\dagger \gamma_R G \gamma_L]}_{t(E)} \]

\[ I_L^{\text{inel}} \sim t_\perp^6 \times \frac{2e}{\hbar} \times \int dE / dE' \ldots \]

- \( -i \gamma_{L(R)} = \Sigma_{L(R)} - \Sigma_{L(R)}^\dagger \)
- \( t(E) \) contains full bath dressing!
- for \( eV \to 0 \) and \( O(t_\perp^2) \sim \) neglect \( I_L^{\text{inel}} \)
Results (qualitative): Low-bias, strong coupling limit

Bath-selfenergy $P(E)$:

$\text{Re } P(E) \sim k_B T$-dependent polaronic manifold

$\text{Im } P(E)$ ("friction") $\sim$ incoherent polaron band, pseudo-gap opens
Results: Transmission and low-bias current

Crossover from tunneling (low T) \(\sim\) activated (high-T) transport
Results: \( t(E_F, T) \) (Arrhenius plot)

Activated behaviour: \( t(E_F) \sim e^{-\text{const.}/k_B T} \)
Results: Scaling of $t(E_F)$ with the chain length $L = N \alpha_{bp}$ ($T=300$ K)

- With increasing coupling to the bath transition from
  
  weak exponential $(\gamma \ll 1)$ \( t_F \sim e^{-\gamma L} \Rightarrow \) algebraic \( t_F \sim L^{-\alpha} \)
Conclusions

• Environment drastically affects charge transport $\sim$
  (i) bath-induced pseudo-gap in the wire electronic spectrum
  (ii) temperature-dependent (incoherent) DOS near $E_F$
  $\sim$ non-zero low-bias current at high $k_B T$
  $\sim$ weak exponential or algebraic $L-$dependence

$\Longrightarrow$ Relation to Xu et al. experiments !?
Outlook

- Interplay with internal dynamical degrees of freedom?
- Sequence complexity?
- Nonequilibrium transport?
Appendix: Landauer theory

Relate charge propagation in a two-terminal setup to an elastic scattering problem \( \sim \) quantum mechanical transmission \( t(E) \)

\[
g = \frac{2e^2}{\hbar} \sum_{\ell=1}^{N} t_{\ell}(E_F)
\]

\( N = \text{number of 1d transport channels} \)

\( t(E) \) can be expressed via Green functions of the scattering region.
Appendix: electron-bath correlator in the strong coupling limit

- $J_0/\omega_c > 1 \sim \text{short-time expansion of bath correlators} \left\langle \mathcal{X}(t)\mathcal{X}^\dagger(0) \right\rangle_B = e^{-\Phi(t)}$

$\Phi(t) \approx it J_0 + (\omega_c t)^2 \kappa_0(T) \sim \text{Gaussian integral} (\hbar = 1)$

$$
P_{\ell j}(z = E + i0^+) = \delta_{\ell j} \int_0^\infty dt \ e^{izt} \ G_{c,\ell\ell}(t) \times \frac{e^{-\Phi(t)}}{\left\langle \mathcal{X}(t)\mathcal{X}^\dagger(0) \right\rangle_B}$$

$$= -i \delta_{\ell j} \sqrt{\pi} \tau_{ph} \ e^{-\frac{\tau_{ph}^2}{2}} x (1 + \text{erf}[i \tau_{ph} z])$$

$$\tau_{ph}^{-2}(T) = \omega_c^2 \kappa_0(T) = \int_0^\infty d\omega \ J(\omega) \ \text{coth} \ \frac{\omega}{2k_B T}$$

$$\tau_{ph}(k_B T \ll \omega_c) \sim (J_0 \omega_c)^{-1/2} \quad \tau_{ph}(k_B T \gg \omega_c) \sim (k_B T J_0)^{-1/2}$$
Appendix: inelastic current for the single-site case

\[ j_{\text{inel}}^L = \frac{2e}{\hbar} t_\perp^2 \gamma_L \gamma_R \int dE \int dE' |G_{12}(E)|^2 |G_{21}(E')|^2 D_{\text{bath}}^{<}(E-E') \times \]
\[ \sim t_\perp^2 \int dE \int dE' |G_{12}(E)|^2 |G_{21}(E')|^2 D_{\text{bath}}^{<}(E-E') \times \]
\[ \times [f_{\text{L}}(E')(1-f_{\text{R}}(E)) - (1-f_{\text{L}}(E)) f_{\text{R}}(E')] \]

\[ D_{\text{bath}}^{<}(E) = \int dt e^{iEt} \langle \mathcal{X}(t) \mathcal{X}^\dagger(0) \rangle_B = \int dt e^{iEt} e^{-\Phi(t)} \]

To order \( O(t_\perp^2) \) and \( eV \rightarrow 0 \) \( \sim \) neglect \( j_{\text{inel}}^L \)
Appendix: behaviour of $P(E, k_B T, J_0/\omega_c)$

$$\det |E 1 - t_\perp^2 \text{Re } P(E) - \mathcal{H}_{\pi-\pi}| = 0, \quad \text{for } J_0 = 0 \rightarrow P(E) \sim 1/E$$

Real $P(E)$

- $T=10$ (black)
- $T=400$ K (red)

E (eV)

Im $P(E)$

- $J_0/\omega_c = 5$ (black)
- $J_0/\omega_c = 12$ (red)