Quantum transport in DNA wires: Influence of a dissipative environment

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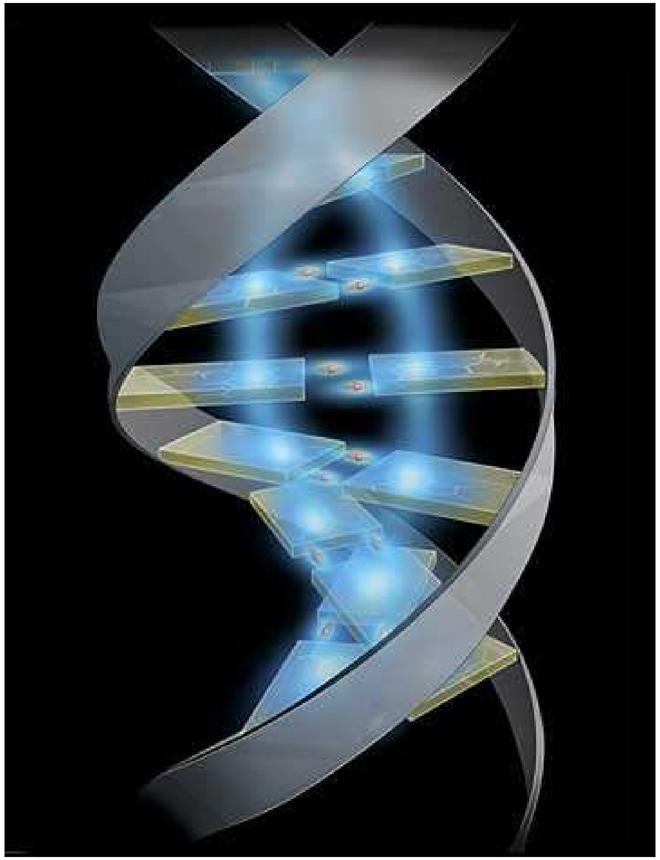
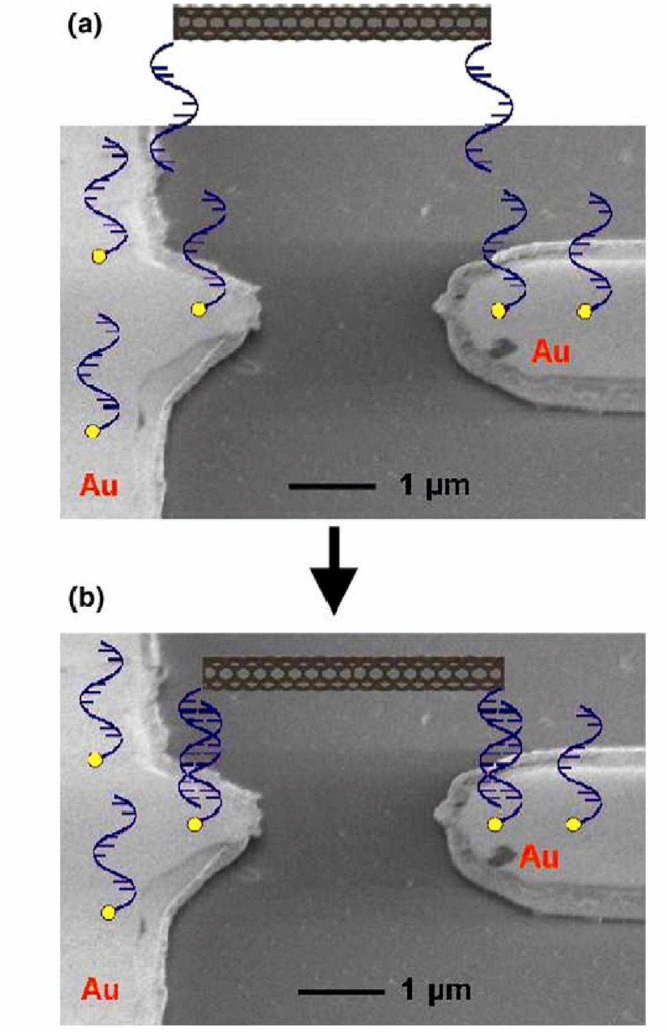


Outline

- Why DNA ?
- Electronic transport in DNA: a bird's eye view
- A model for a dissipative DNA wire
- Formalism and approximations
- Strong dissipative limit
- Conclusions

Why DNA ?



M. Hazani *et al.* , Chem. Phys. Lett. (2004)

- Groundbreaking : repair of oxidative damage
  $\rightsquigarrow$ ET over long distances (~ 40 Å)
  (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))

Electronic transport in DNA: a bird's eye view

- Experiments: DNA is insulator, metal, semiconductor

  $\rightsquigarrow$ sample preparation and experimental conditions are crucial

  (dry vs. aqueous environment, metal-molecule contacts, single molecules vs. bundles $\cdots$)

- Theory: Variety of factors modifying charge propagation:

  static disorder, dynamical disorder, environment (hydration shell, counterions)

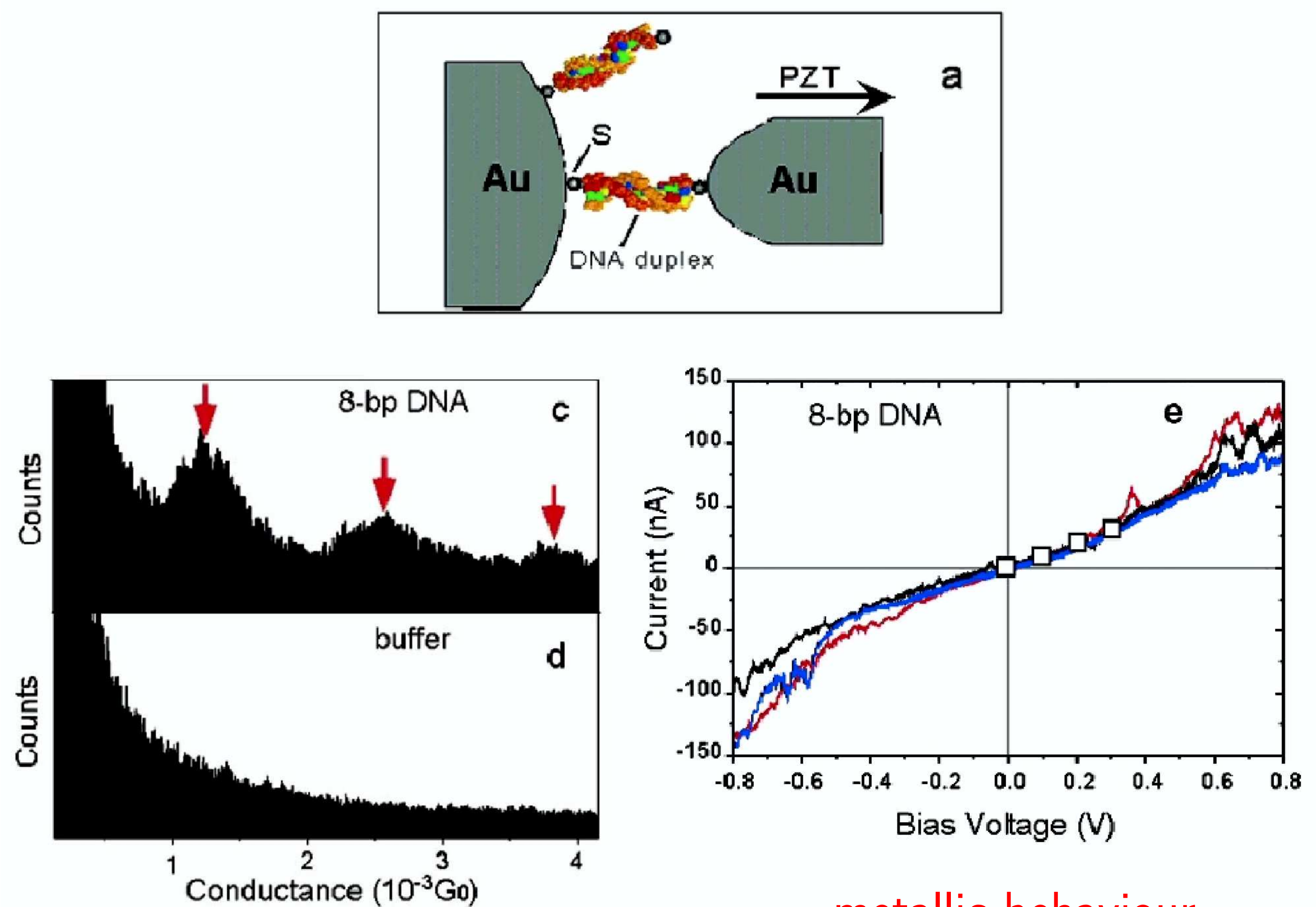
see: D. Porath, G. Cuniberti, and R. Di Felice,

Charge Transport in DNA-Based Devices

Top. Curr. Chem. (2004)

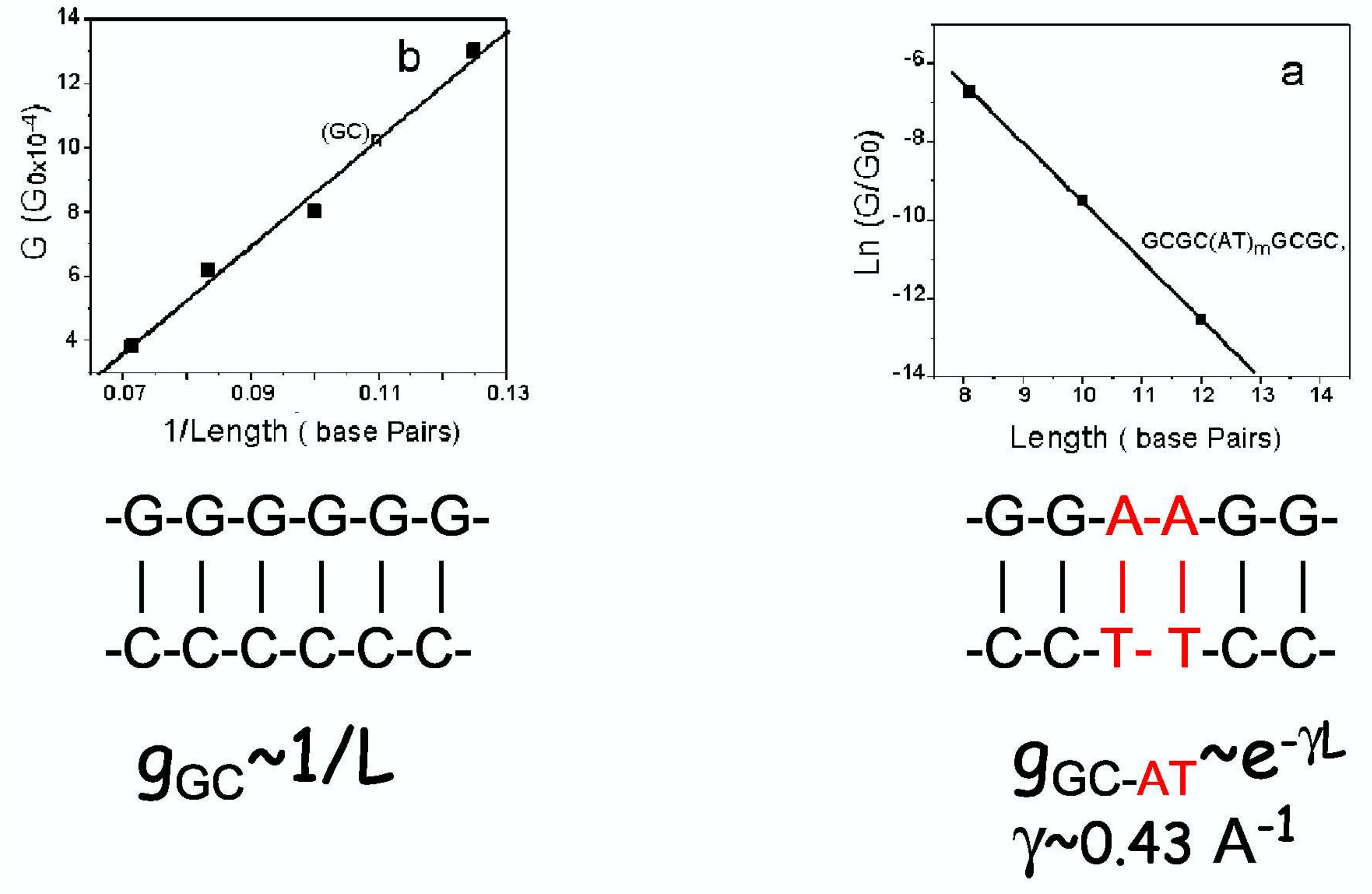
Transport in *single* Poly(GC) oligomers in *water*

B. Xu *et al.* Nanoletters 4, 1105 (2004)



metallic behaviour

Transport in *single* Poly(GC) oligomers in *water*

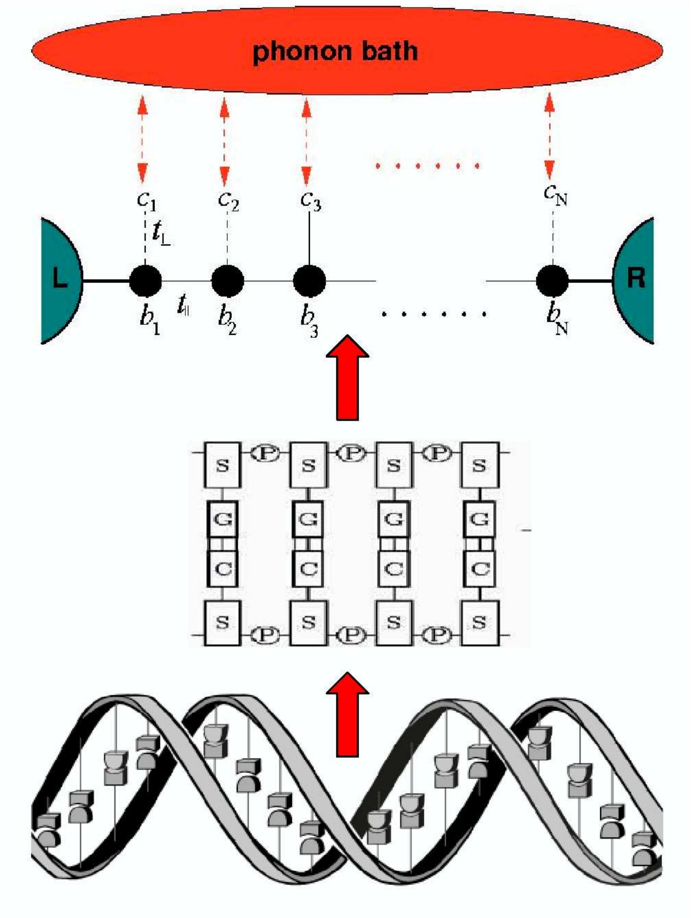


Ab initio (H. Wang *et al.* PRL (2004)): dry Poly(GC) $\rightsquigarrow e^{-\gamma L}, \gamma \sim 1.5$ Å^{-1}

Algebraic behaviour induced by the environment ?

A model for a dissipative DNA wire

- *ab initio* $\rightsquigarrow$ (i) decoupled HOMO/LUMO channels, (ii) backbones non conducting, (iii) band gap $\sim$ 2 eV (dry), but modified by water shell+counterions

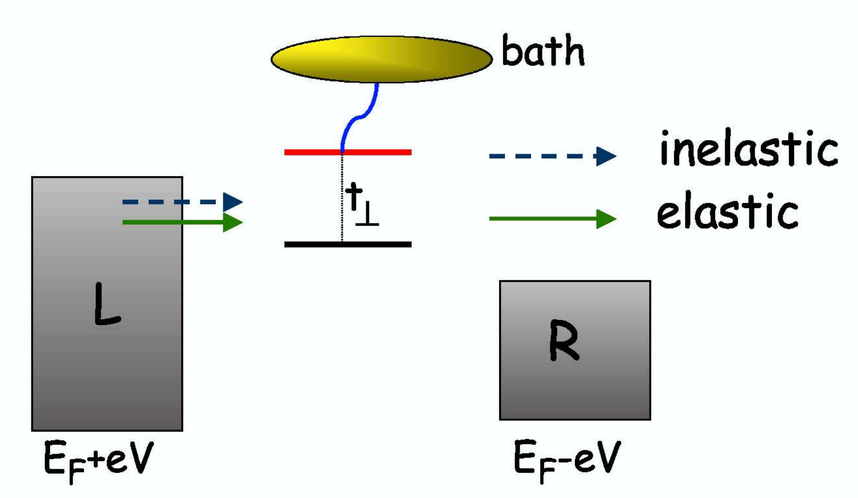


$$
\begin{aligned}
\mathcal{H} \;=\; & \underbrace{\sum_j \epsilon_{b,j} b_j^\dagger b_j - t_{||} \sum_{<i,j>} \left(b_i^\dagger b_j + \mathrm{H.c.}\right)}_{\mathcal{H}_\mathcal{C}} + \underbrace{\sum_j \epsilon_j c_j^\dagger c_j}_{\mathcal{H}_\mathrm{c}} \\
& - \underbrace{t_\perp \sum_j \left(b_j^\dagger c_j + \mathrm{H.c.}\right)}_{\mathcal{H}_{\mathcal{C}-\mathrm{c}}} + \underbrace{\sum_{\mathbf{k}\in \mathrm{L,R},\sigma} \epsilon_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma}^\dagger d_{\mathbf{k}\sigma}}_{\mathcal{H}_{\mathcal{L}/\mathcal{R}}} \\
& + \underbrace{\sum_{\mathbf{k}\in \mathrm{L},\sigma} \left(V_{\mathbf{k},1}\, d_{\mathbf{k}\sigma}^\dagger\, b_1 + \mathrm{H.c.}\right) + \sum_{\mathbf{k}\in \mathrm{R},\sigma} \left(V_{\mathbf{k},N}\, d_{\mathbf{k}\sigma}^\dagger\, b_N + \mathrm{H.c.}\right)}_{\mathcal{H}_{\mathcal{L}/\mathcal{R}-\mathcal{C}}} \\
& + \underbrace{\sum_\alpha \Omega_\alpha B_\alpha^\dagger B_\alpha}_{\mathcal{H}_\mathcal{B}} + \underbrace{\sum_{\alpha,j} \lambda_\alpha c_j^\dagger c_j \left(B_\alpha + B_\alpha^\dagger\right)}_{\mathcal{H}_{\mathcal{F}-\mathcal{B}}} .
\end{aligned}
$$

Formalism and approximations

- Green function techniques

- Phonon bath $\rightsquigarrow$ continuous frequency spectrum $J(\omega) \sim (\frac{\omega}{\omega_c})^s$



- Low-bias limit

  $\rightsquigarrow$ neglect nonequilibrium effects

  $\rightsquigarrow$ no real electron-phonon processes

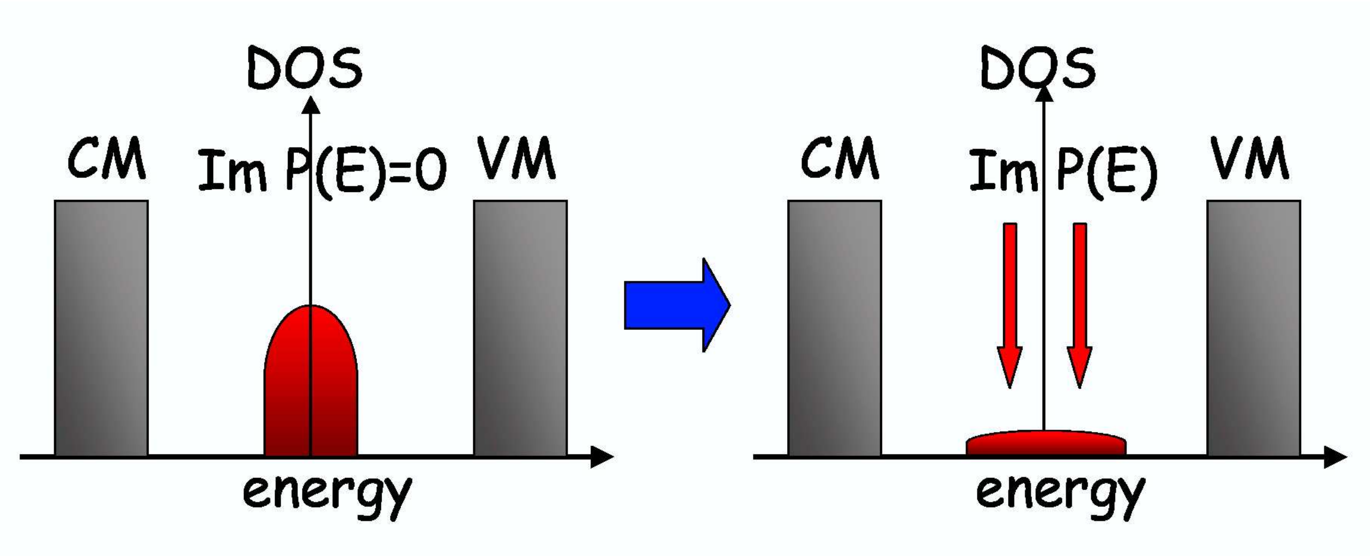
- Landauer-like expression :

  $t(E) = Tr[G^\dagger \Gamma_R \, G \, \Gamma_L]$

  $t(E)$ includes virtual electron-phonon processes

Low-bias, strong dissipative limit

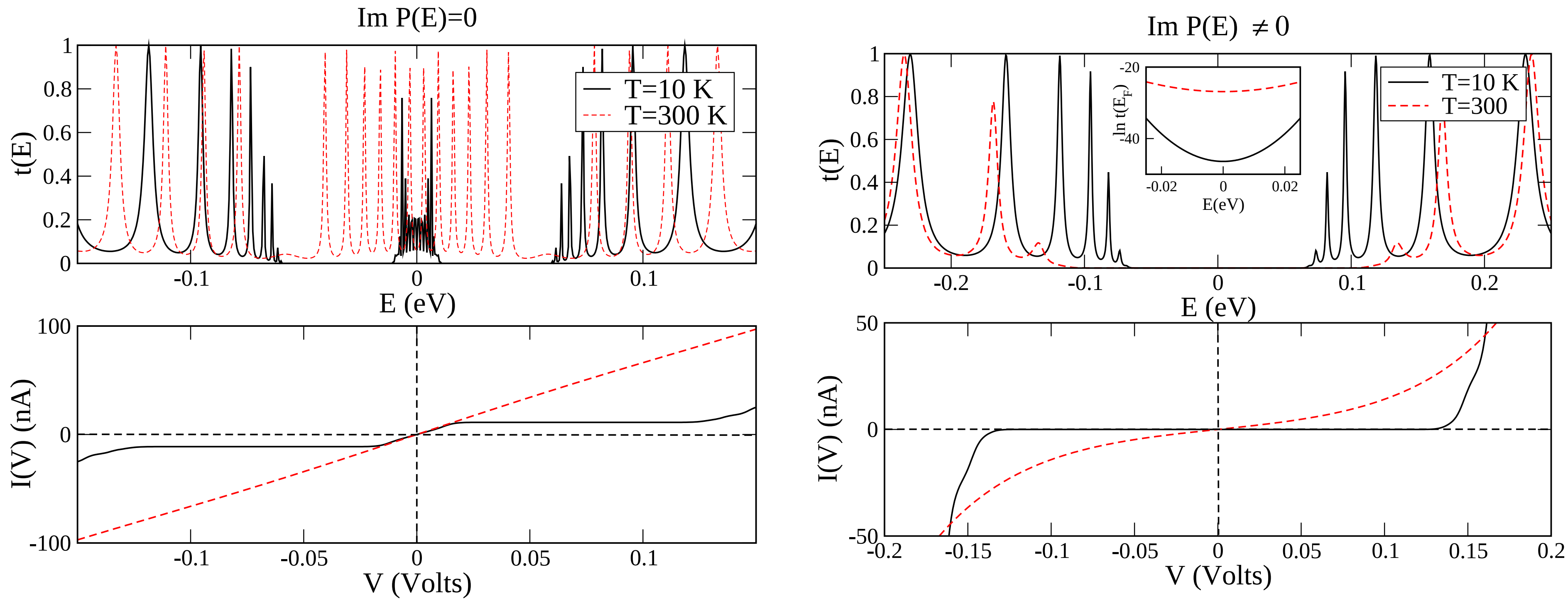
New $k_B T$-dependent electronic manifold around $E_F = 0$



Im P(E) ("bath-friction") strongly suppresses the central manifold

incoherent polaronic band $\rightsquigarrow$ pseudo-gap opening

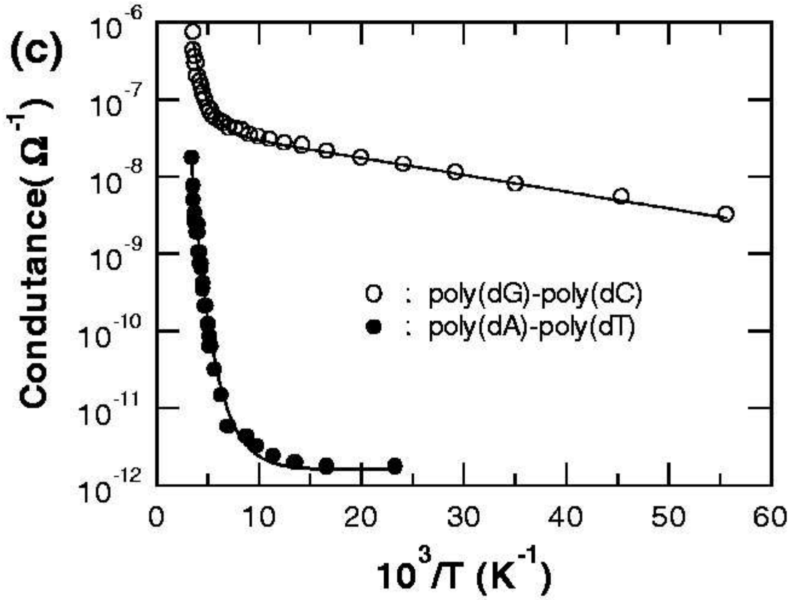
Transmission t(E) and low-bias current I(V)

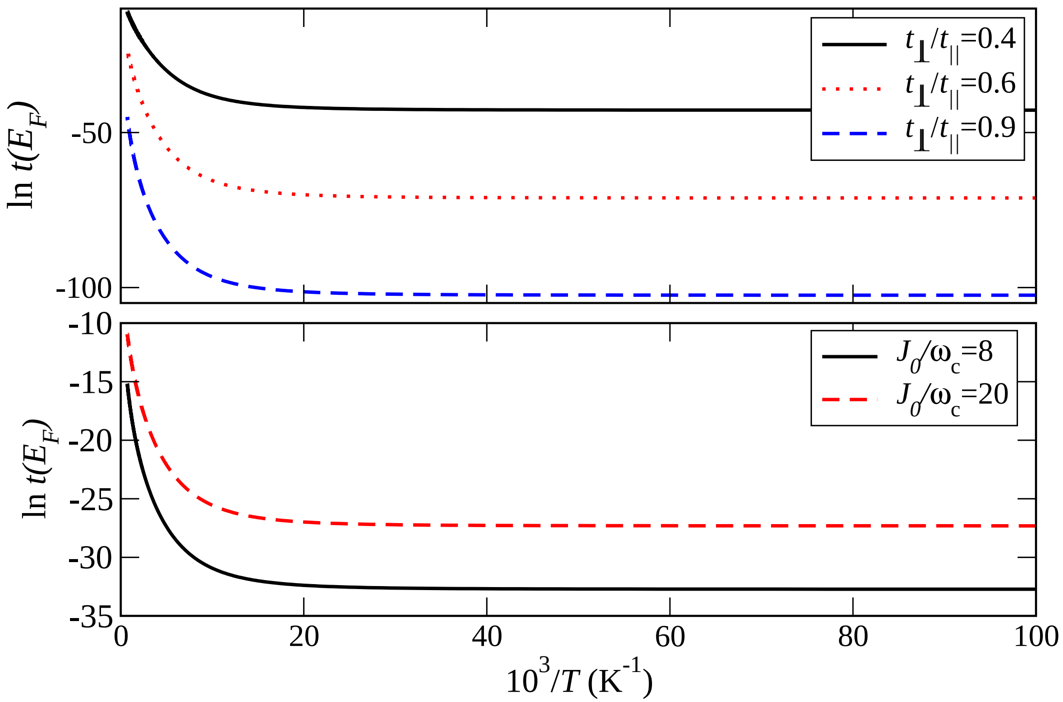


- pseudo-gap increases with temperature
- central band DOS also increases with temperature
- $I(V)$ displays "metallic" behaviour at high $k_\mathrm{B}T$

Temperature dependence of $t(E_F)$ (Arrhenius plot)

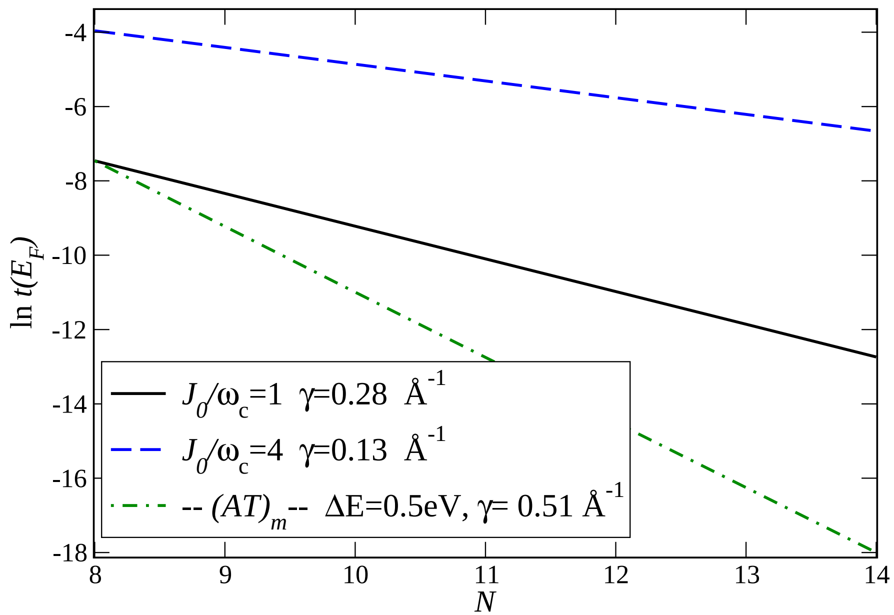
Yoo et al. PRL 87, 198102 (2001)

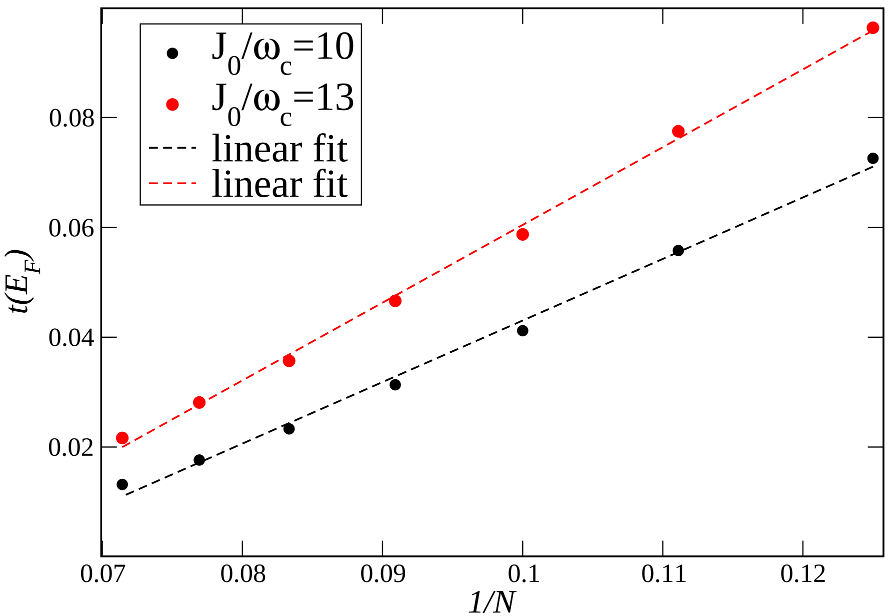




Activated behaviour

Scaling of $t(E_F)$ with the chain length $L = Na_{\text{bp}}$





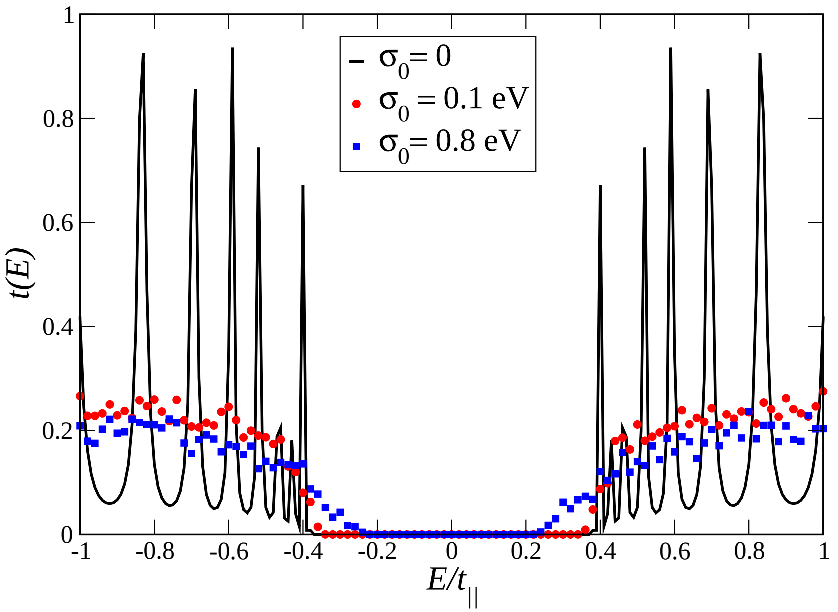
- Increasing coupling to the bath

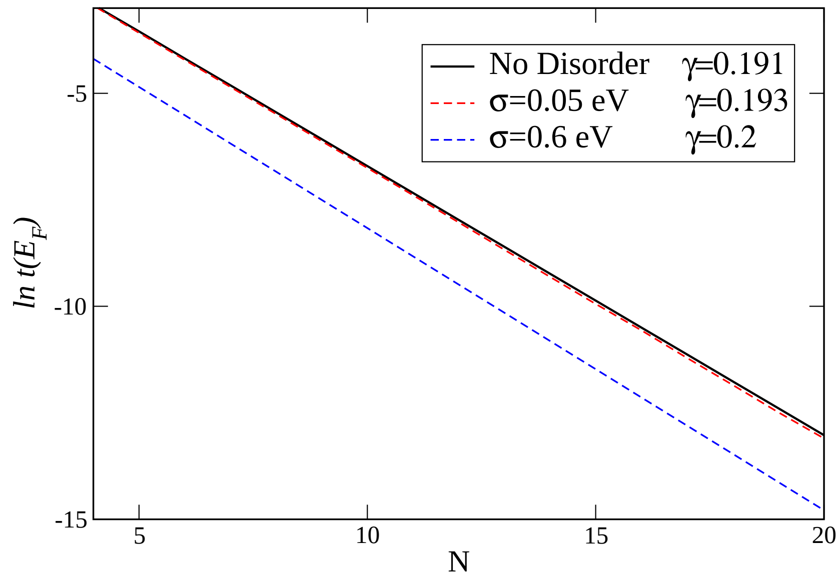
exponential $t_F \sim e^{-\gamma L} \Longrightarrow$ algebraic $t_F \sim L^{-\alpha}$ dependence

- Exponential dependence is not related to virtual tunneling through a gap ($\gamma \ll 1$ Å^{-1})
- Introduction of a barrier $\leadsto$ (AT)$_n$ pairs, enforces exp-dependence

Structural base-pairs fluctuations

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





Disorder does not appreciably affect the pseudo-gap formation

Conclusions

- Environment drastically affects charge transport
- Strong dissipative regime:

  (i) bath-induced pseudo-gap

  (ii) finite k_BT-dependent DOS near $E_F \rightsquigarrow$

  activated behaviour

  weak exponential or algebraic $L-$dependence

- Contact to Xue *et al.* experiments

  (i) large currents $\sim 50 - 200\,nA$

  (ii) (bath-induced) algebraic L-dependence

  (iii) $\gamma[(\mathrm{AT})_n] \approx 0.15$ Å$^{-1} < \gamma^{\mathsf{Xue}}[(\mathrm{AT})_\mathrm{n}] = 0.43$ Å^{-1}

Perspectives