Quantum transport in DNA wires: Influence of a dissipative environment

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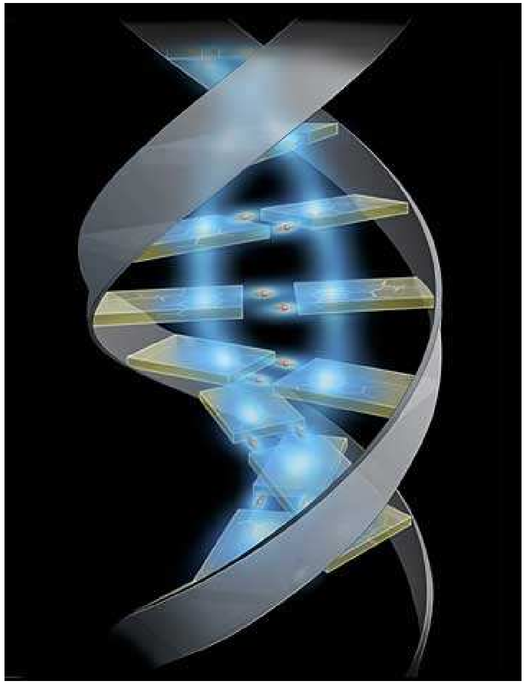
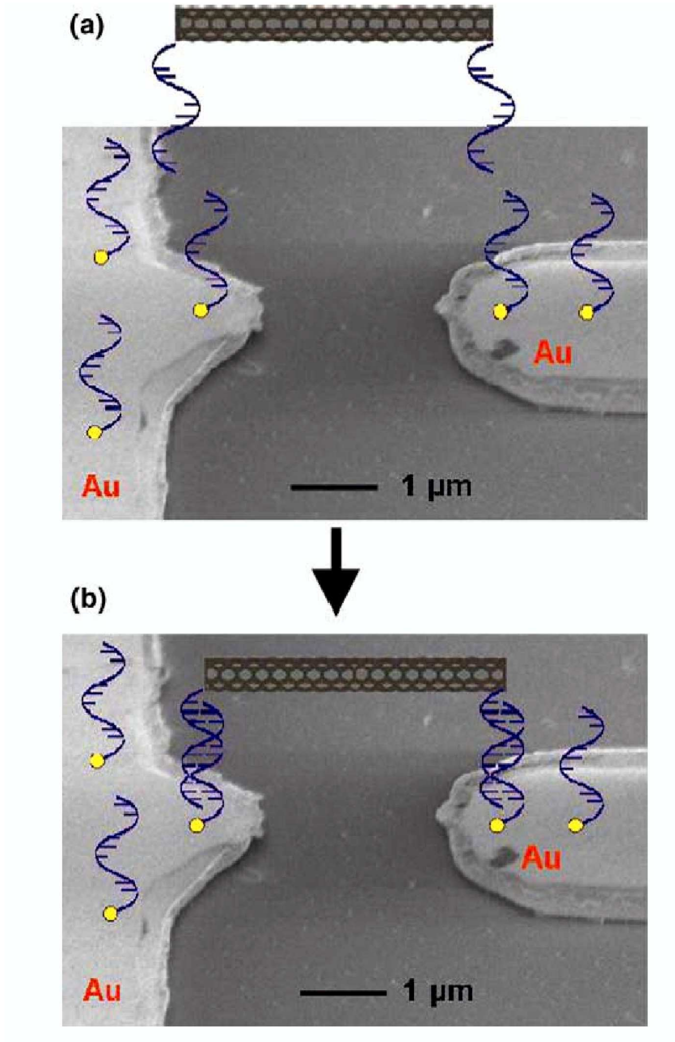


In cooperation with S. Mandal and G. Cuniberti

Outline

- Why DNA ?
- Electronic transport in DNA: a bird's eye view
- A model for a dissipative DNA wire
- Electronic transport and Green functions
- Results for strong dissipation (wet DNA)
- Conclusions

Why DNA ?



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

- Groundbreaking : repair of oxidative damage

  $\rightsquigarrow$ ET over long distances ($\sim 40\,\mathring{A}$)

  (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))

  (in Dresden W. Pompe/M. Mertig !)

Electronic transport in DNA: a bird's eye view

- 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)

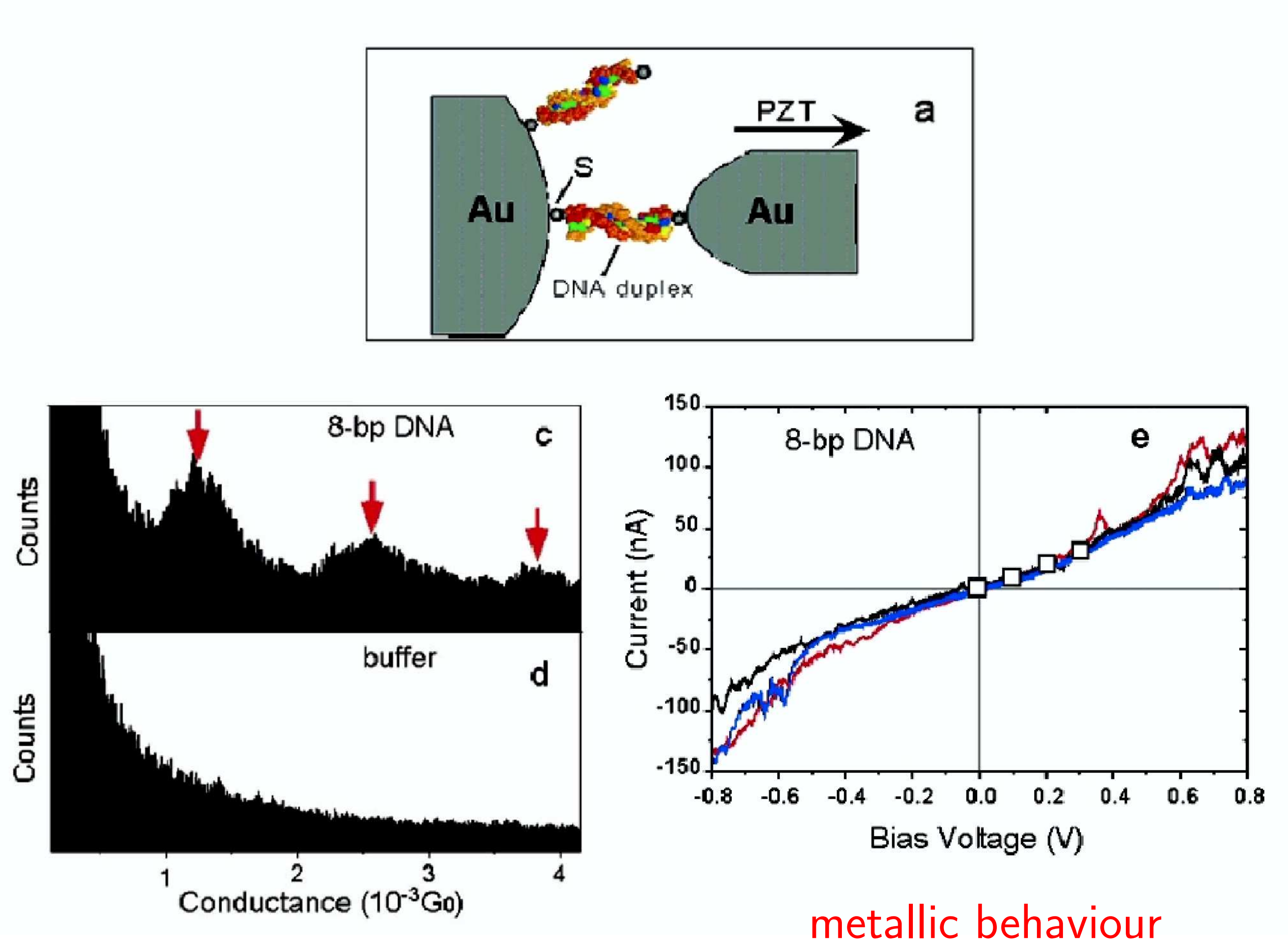
see: D. Porath, G. Cuniberti, and R. Di Felice,

Charge Transport in DNA-Based Devices

Top. Curr. Chem. (2004)

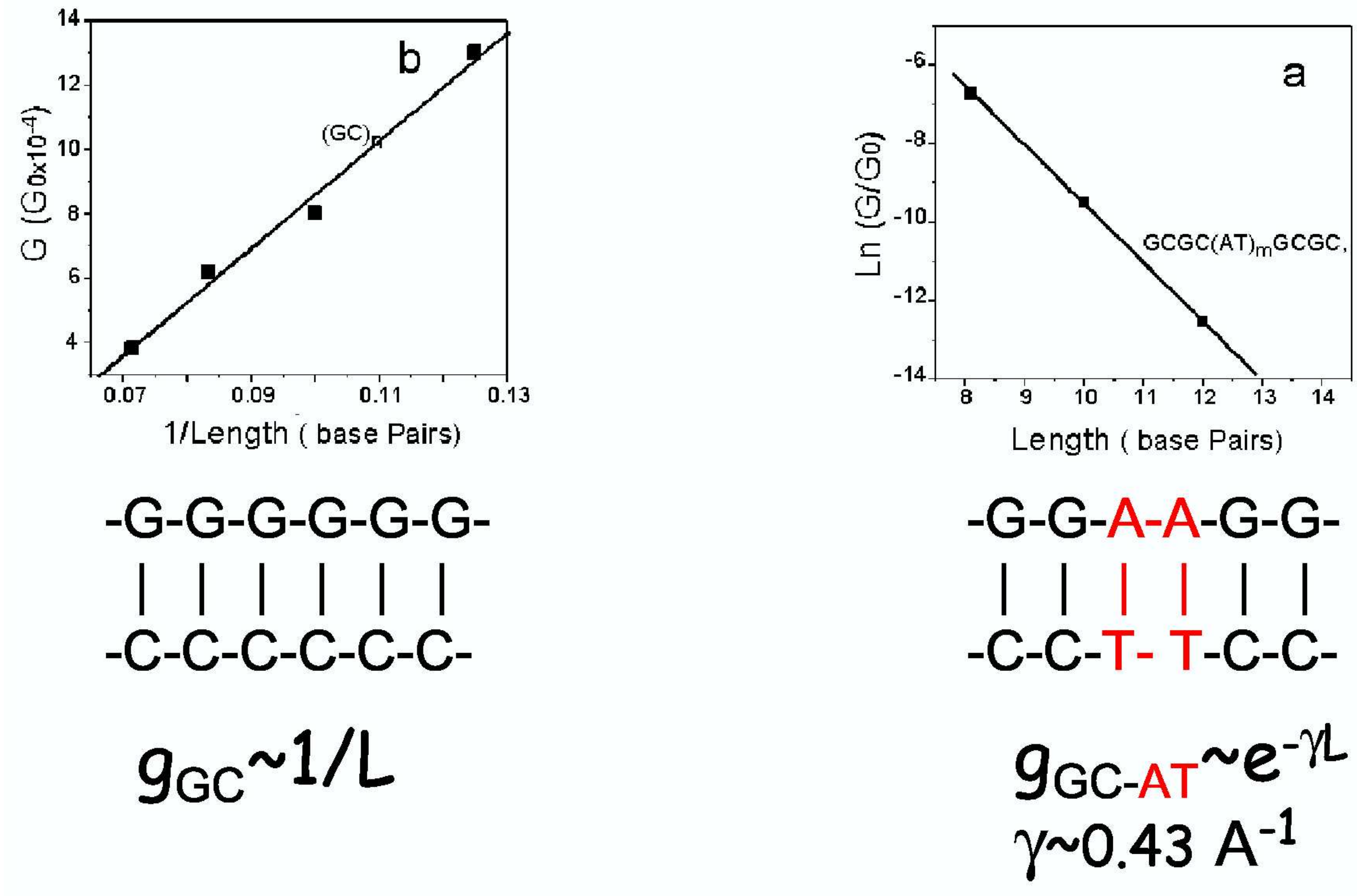
Transport in *single* Poly(GC) oligomers in *water*

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



metallic behaviour

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

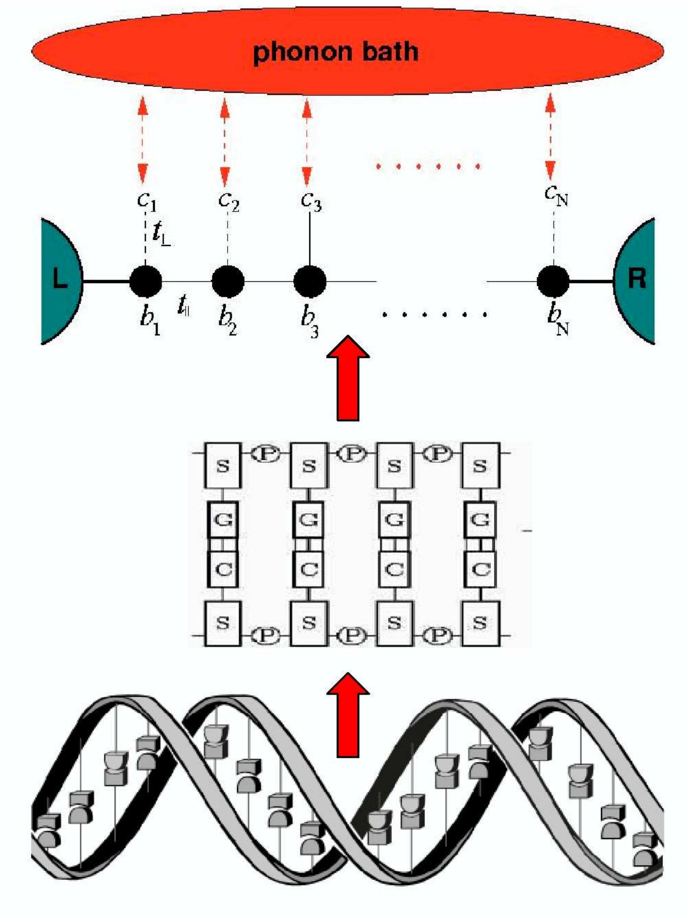


Ab initio (H. Wang *et al.* PRL (2004)): dry Poly(GC) $\rightsquigarrow e^{-\gamma L}, \gamma \sim 1.5$ Å^{-1}

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 reduced by water shell+counterions



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

Green function techniques

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

$$\mathcal{H}_c \rightarrow \sum_j (\epsilon_j + \underbrace{\sum_\alpha \frac{\lambda_\alpha^2}{\Omega_\alpha}}_{\Delta}) c_j^\dagger c_j \qquad \mathcal{H}_{\mathcal{C}-c} \rightarrow -t_\perp \sum_j [\, b_j^\dagger\, c_j\, \underbrace{\exp\left(-\sum_\alpha \frac{\lambda_\alpha}{\Omega_\alpha}(B_\alpha^\dagger - B_\alpha)\right)}_{\mathcal{X}} + \mathrm{H.\ c.}\,]$$

- Green functions ($\hbar = 1$)

$$G_{jl}(t) \;=\; -\mathrm{i} < \mathcal{T}\left\{\mathrm{b_j(t)}, \mathrm{b}_\ell^\dagger(0)\right\} >$$

$$\mathbf{G}^{-1}(E) \;=\; E\mathbf{1} - \mathcal{H}_\mathcal{C} - \Sigma_{L/R}(E) - \; t_\perp^2 \mathbf{P}(E)$$

$$P_{\ell j}(E) \;=\; -\mathrm{i}\, \delta_{\ell \mathrm{j}} \int_0^\infty \mathrm{dt}\, \mathrm{e}^{\,\mathrm{i}\,(E+\mathrm{i}\,0^+)\mathrm{t}}\, \mathrm{G_c(t)} \times \underbrace{\mathrm{e}^{\,-\Phi(t)}}_{\left\langle \mathcal{X}(\mathrm{t}) \mathcal{X}^\dagger(0) \right\rangle_\mathrm{B}}$$

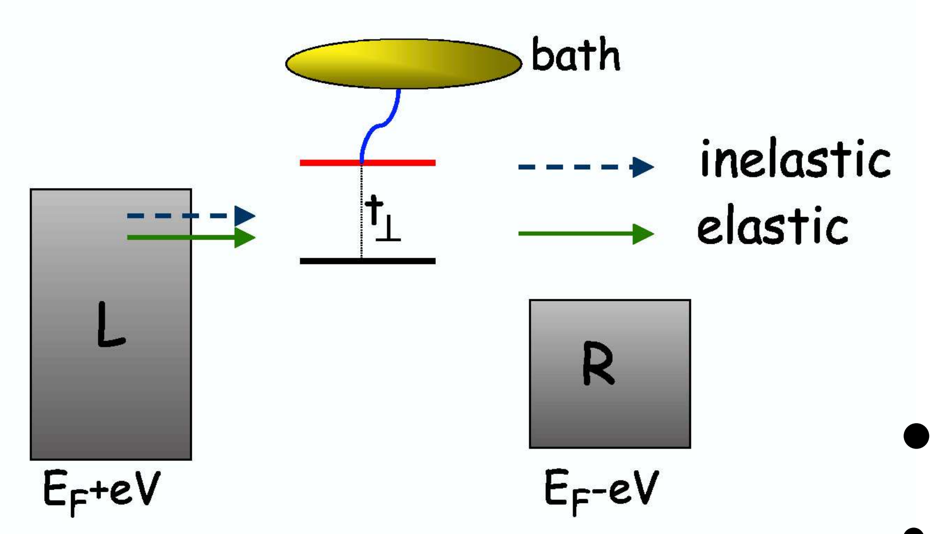
- continuous bath frequency distribution ($N \rightarrow \infty$) $\rightsquigarrow$ spectral density :

$$J(\omega) \;=\; \sum_\alpha \lambda_\alpha^2 \delta(\omega - \Omega_\alpha) = J_0 (\frac{\omega}{\omega_c})^s e^{-\omega/\omega_c} \theta(\omega),$$

$$\Phi(t) \;=\; \int_0^\infty \mathrm{d}\omega\, \frac{J(\omega)}{\omega^2} \left[(\mathrm{N}(\omega) + 1)(1 - \mathrm{e}^{-\mathrm{i}\,\omega \mathrm{t}}) + \mathrm{N}(\omega)(1 - \mathrm{e}^{\mathrm{i}\,\omega \mathrm{t}}) \right]$$

The current

$$I_{\rm el} = \frac{2e}{h} \int dE\,(f_L - f_R)\, t(E)$$
$$= \frac{2e}{h} \Gamma_{\rm L}\,\Gamma_{\rm R} \int dE\,(f_L - f_R)\, |G(E)|^2$$

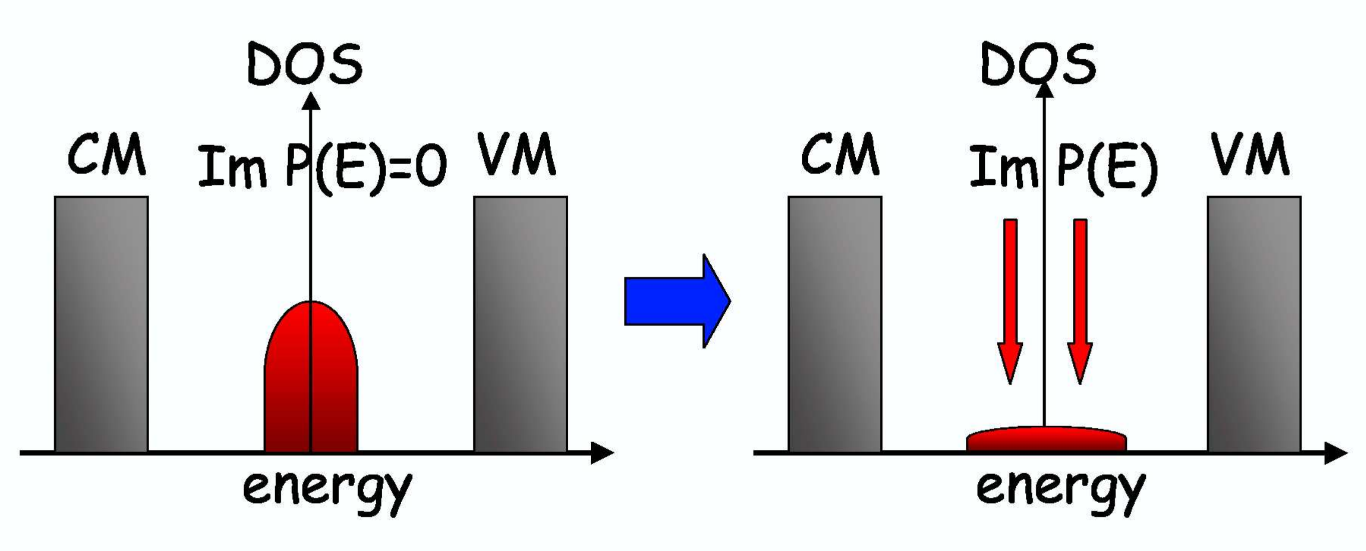


$$I_{\rm inel} \sim \Gamma_{\rm L}\,\Gamma_{\rm R}\, t_\perp^4 \times \sum_\alpha \lambda_\alpha^2 (\cdots)$$

- For $\lambda_\alpha \to 0$, $I_{\rm el} \to$ Landauer formula
- G(E) contains all contributions from the bath !
- for $eV \to 0$ and to lowest order in $t_\perp \rightsquigarrow$ neglect $I_{\rm inel}$
- focuse on $t(E) \rightsquigarrow$ modifications of the electronic spectrum induced by the bath

DOS in the strong dissipative limit:

New k_BT-dependent electronic manifold around $E_F=0$

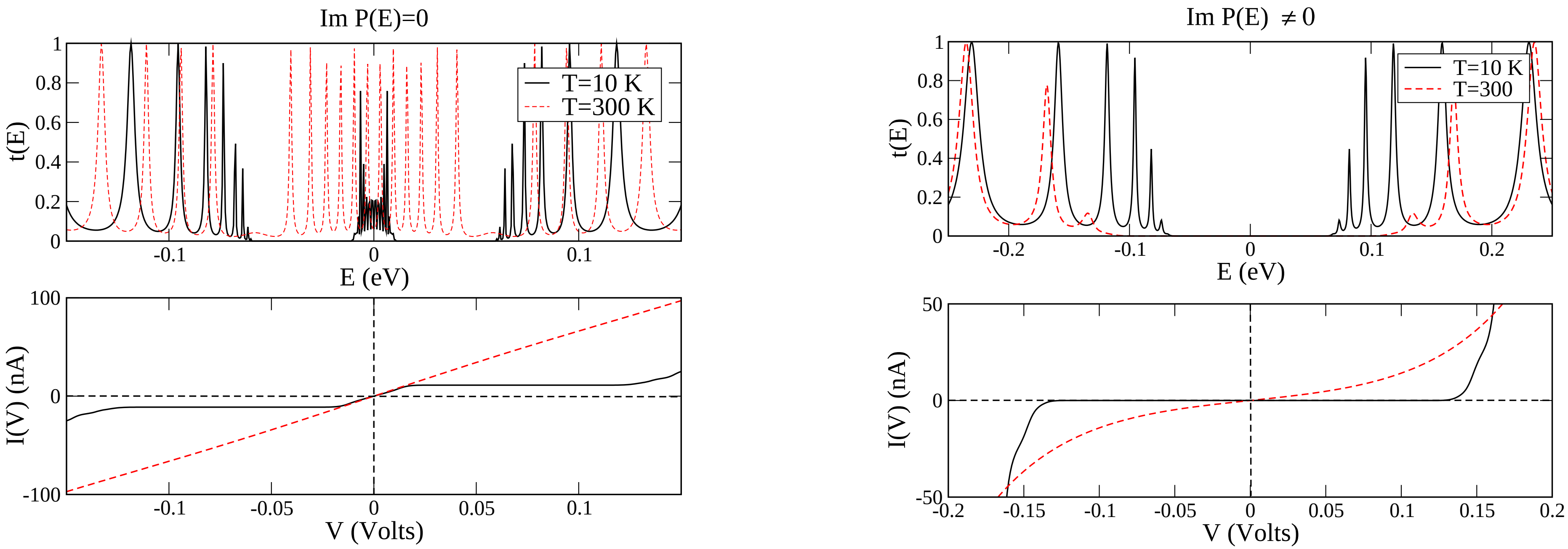


Im P(E) ("bath-friction") strongly suppresses the central manifold

incoherent polaronic band $\rightsquigarrow$ pseudo-gap opening

$$G(E \sim E_{\mathsf{F}}) \sim \underbrace{\frac{t_\perp^2 \, e^{-\kappa_0(T)}}{E - (\epsilon + \Delta) + \mathrm{i}\,\eta}}_{\text{coherent part}} + t_\perp^2 \, e^{-\kappa_0(T)} \Sigma^{(2)}(E) + \cdots \text{(all orders)}$$

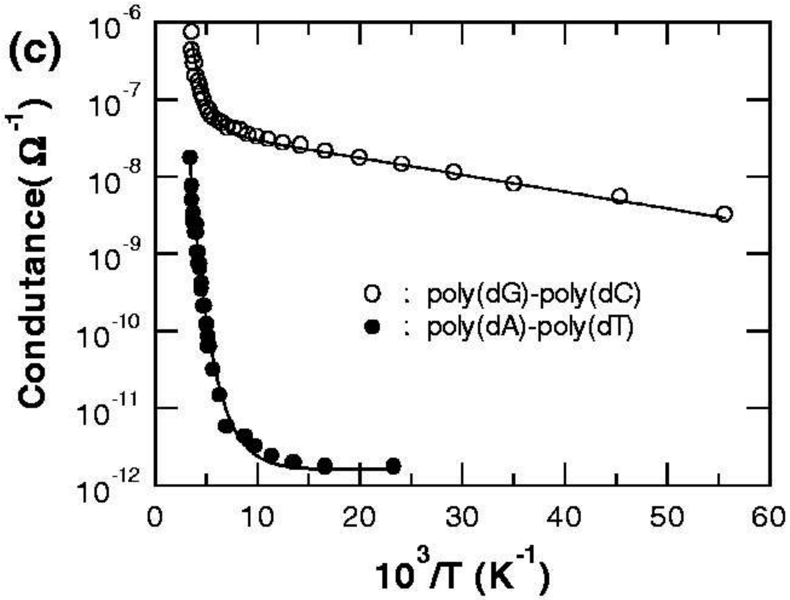
Transmission t(E) and low-bias current I(V)

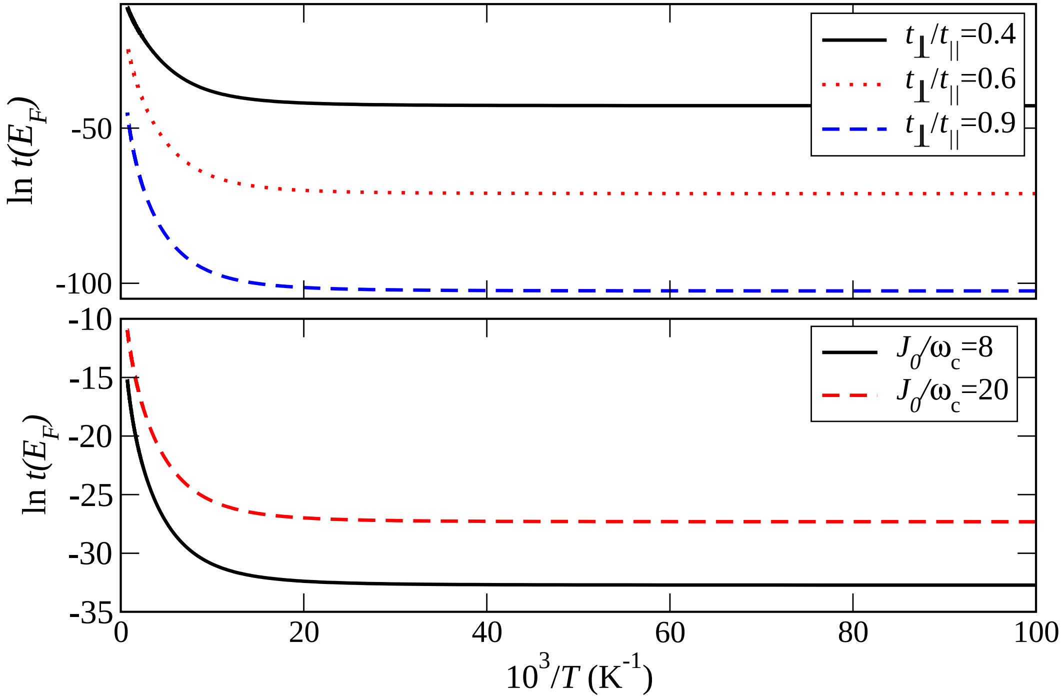


- pseudo-gap increases with temperature
- central band DOS also increases with temperature
- $I(V)$ displays linear behaviour at high $k_{\mathrm{B}}T$

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

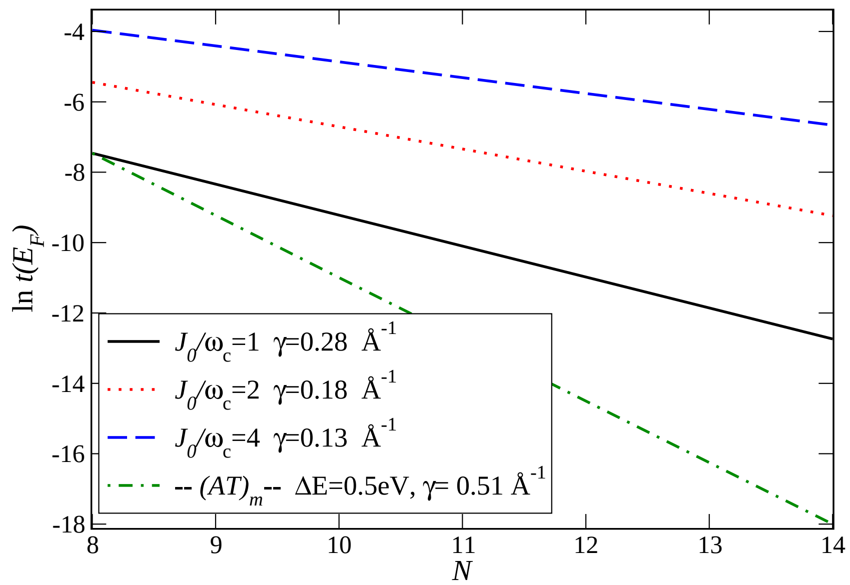
Yoo et al. PRL 87, 198102 (2001)

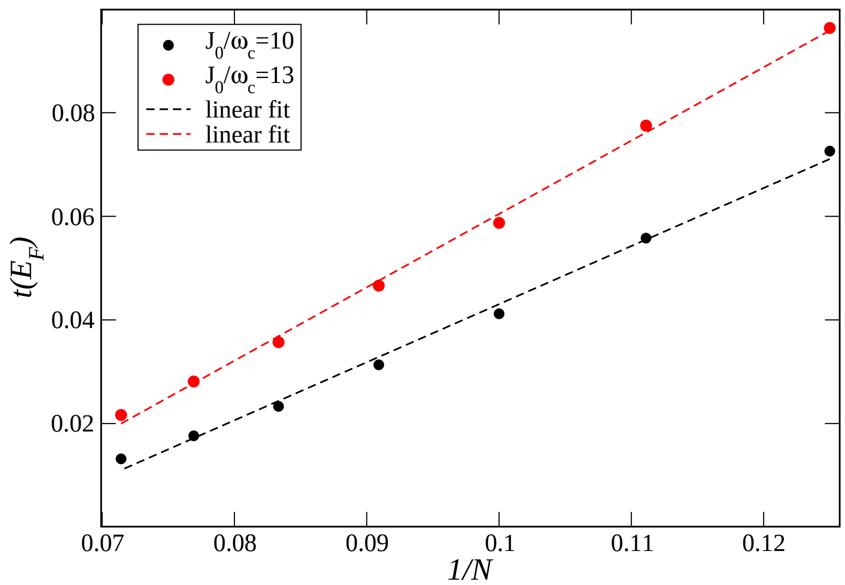




Activated behaviour

Scaling of $t(E = E_F)$ with the chain length N





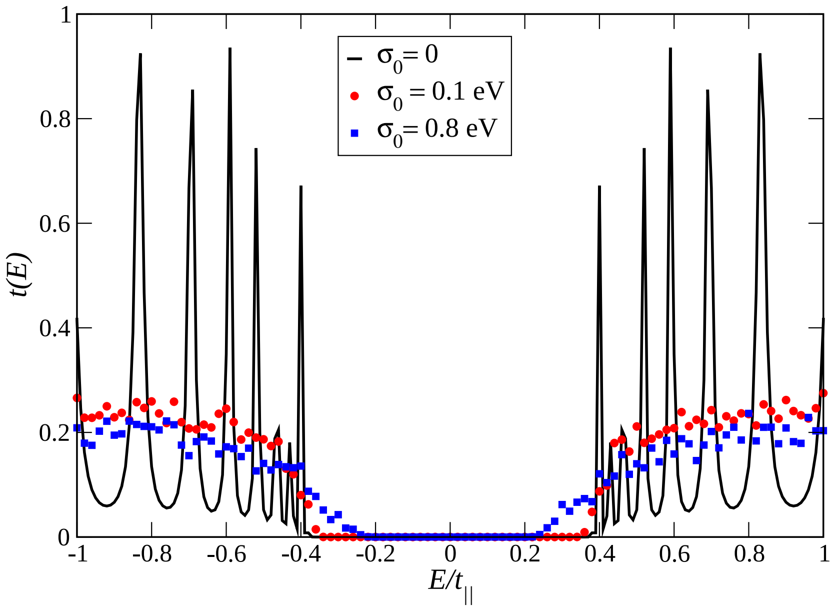
- Increasing coupling to the bath

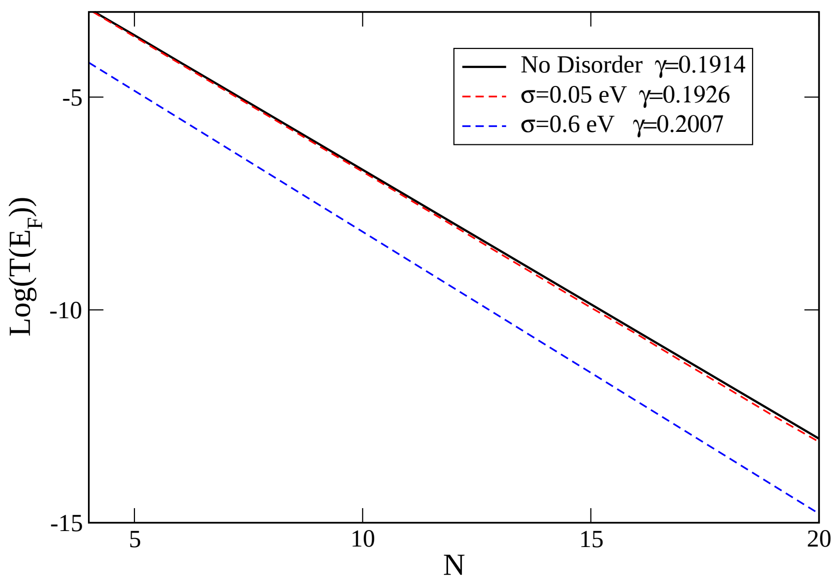
$$\text{exponential } t_F \sim e^{-\gamma L} \Longrightarrow \text{algebraic } t_F \sim L^{-\alpha} \quad \text{dependence } (L = N a_0)$$

- Exponential dependence is not related to virtual tunneling through a gap ($\gamma \ll 1$ Å^{-1})
- Introduction of a barrier $\rightsquigarrow$ (AT)$_n$ pairs, enforces exp-dependence, $\gamma \sim 0.5$ Å^{-1}

Structural 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 and weak exponential

  $L-$dependence of $t(E_F)$

- Contact to Xue *et al.* experiments

  (i) large currents $\sim 50 - 200\, nA \rightsquigarrow$ not problem

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

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

Perspectives