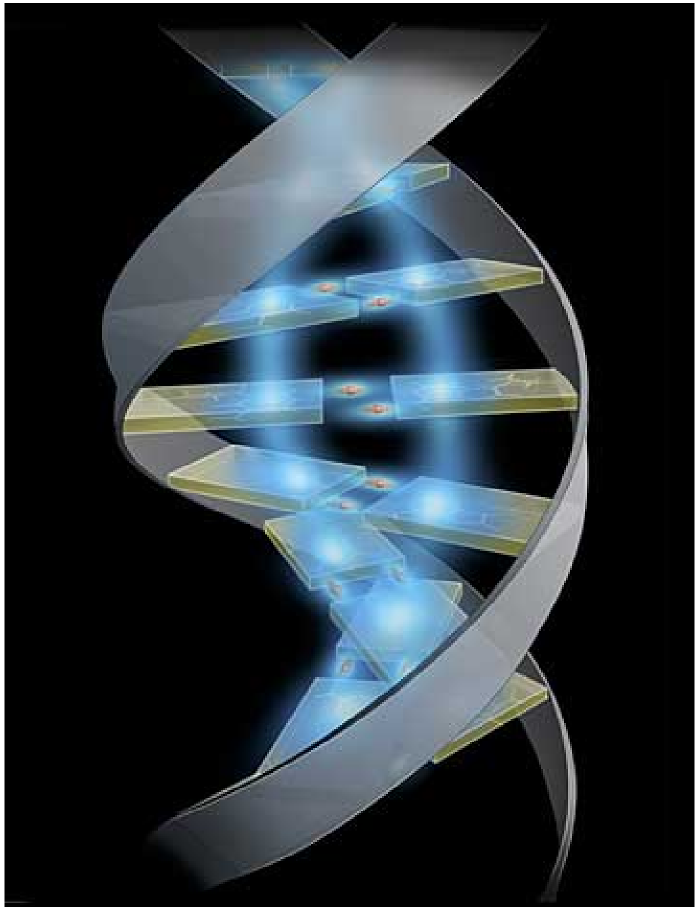
Quantum transport in DNA wires: Influence of a dissipative environment



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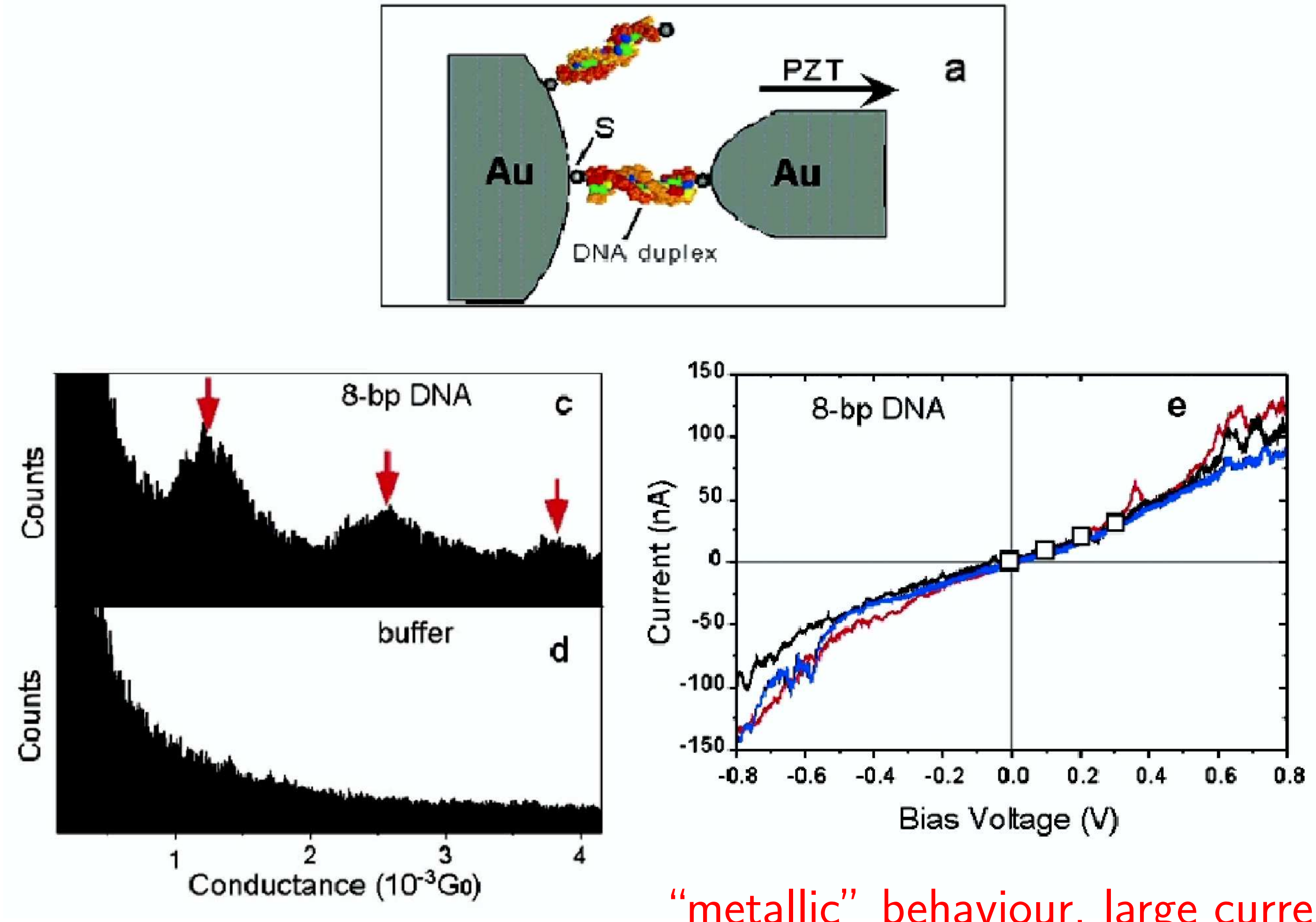
Outline

- Tao's experiment (Nano Letters 4, 1105 (2004))
- A minimal model for DNA wires in solution
- Results: low-bias, strong coupling limit
- Conclusions and Outlook

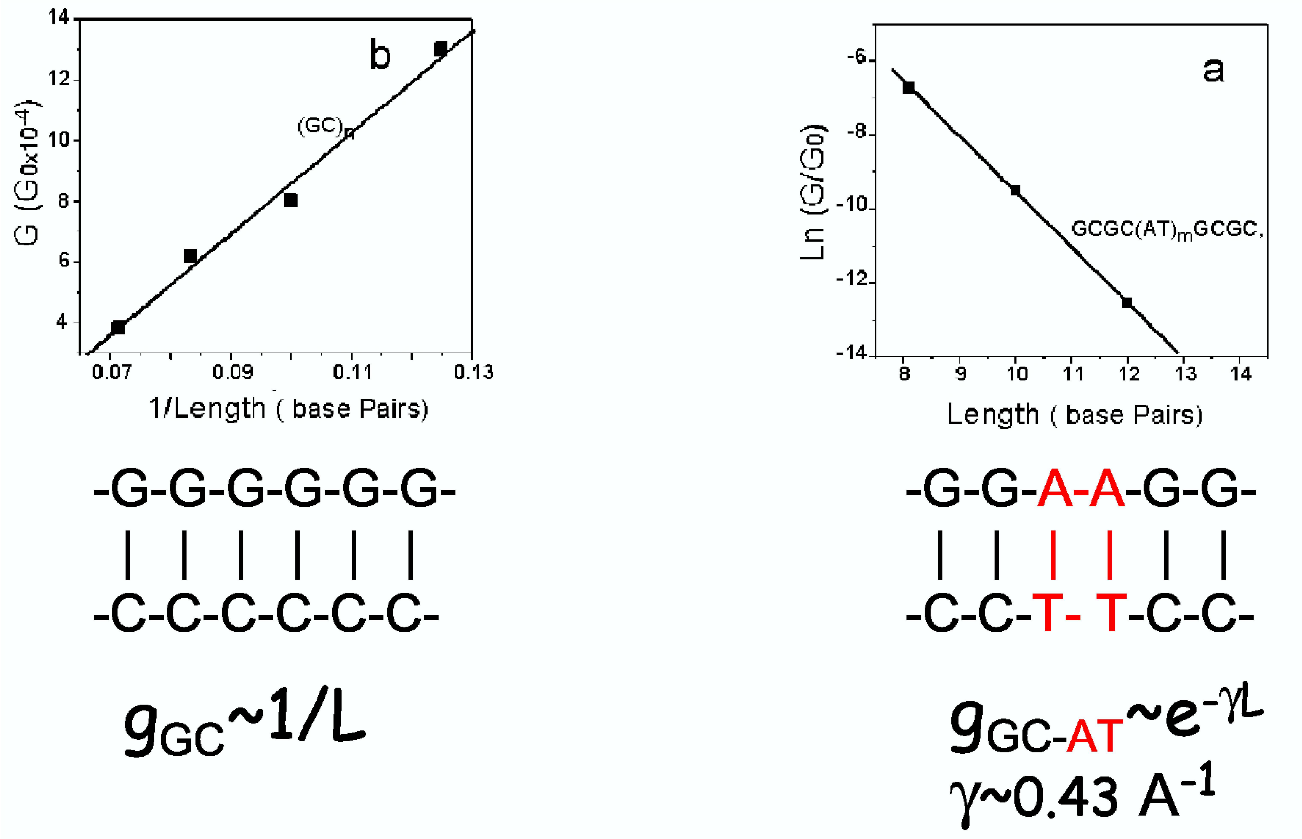
Motivation: Transport in *single* Poly(GC) oligomers in *water* (I)

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



"metallic" behaviour, large currents

Motivation: Transport in *single* Poly(GC) oligomers in *water* (II)

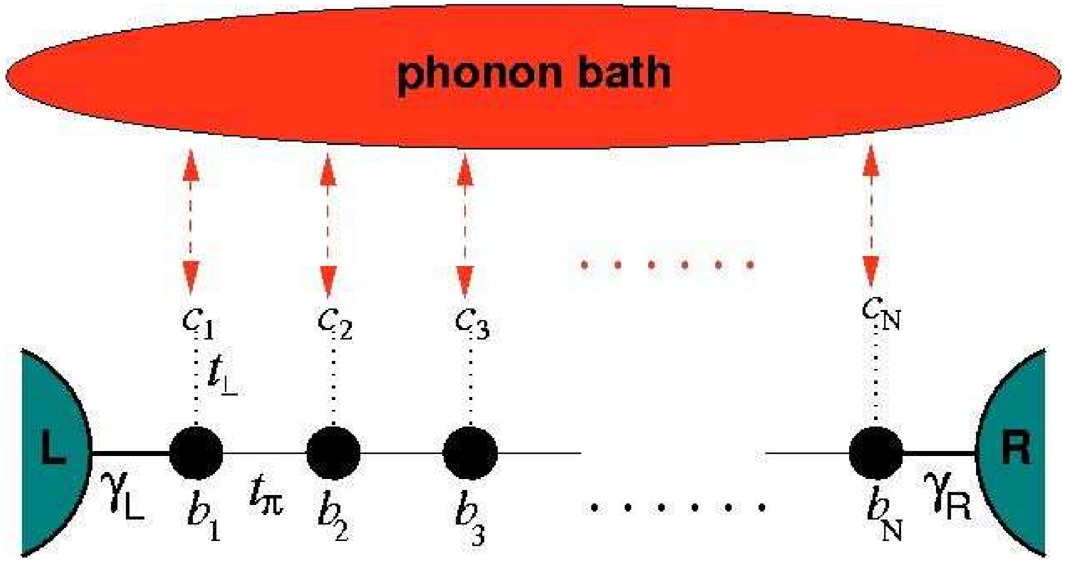


but ... *Ab initio* (H. Wang *et al.* (2004)): dry Poly(GC) $\rightsquigarrow e^{-\gamma L}, \gamma \sim 1.5$ Å^{-1}

$\Longrightarrow$ Influence of the environment ?

A minimal model for a DNA wire in "water"

- Low-bias $\rightsquigarrow$ only frontier orbitals relevant (HOMO/LUMO$\rightsquigarrow$ π character)
- HOMO/LUMO-charge density on G/C-bases $\rightsquigarrow$ 1D channels
- Backbones do not contribute to transport $\rightsquigarrow$ perturbation of π-stack $\rightsquigarrow$ gap opening



A minimal model for a DNA wire in "water"

"Dry" model: G. Cuniberti et al. PRB **65** 241314(R) (2002)

$$\mathcal{H} = \overbrace{\sum_j \epsilon_{b,j} b_j^\dagger b_j - t_\pi \sum_j \left(b_j^\dagger b_{j+1} + \mathrm{H.c.} \right)}^{\text{HOMO(LUMO)}\rightsquigarrow \mathcal{H}_{\pi-\pi}}$$

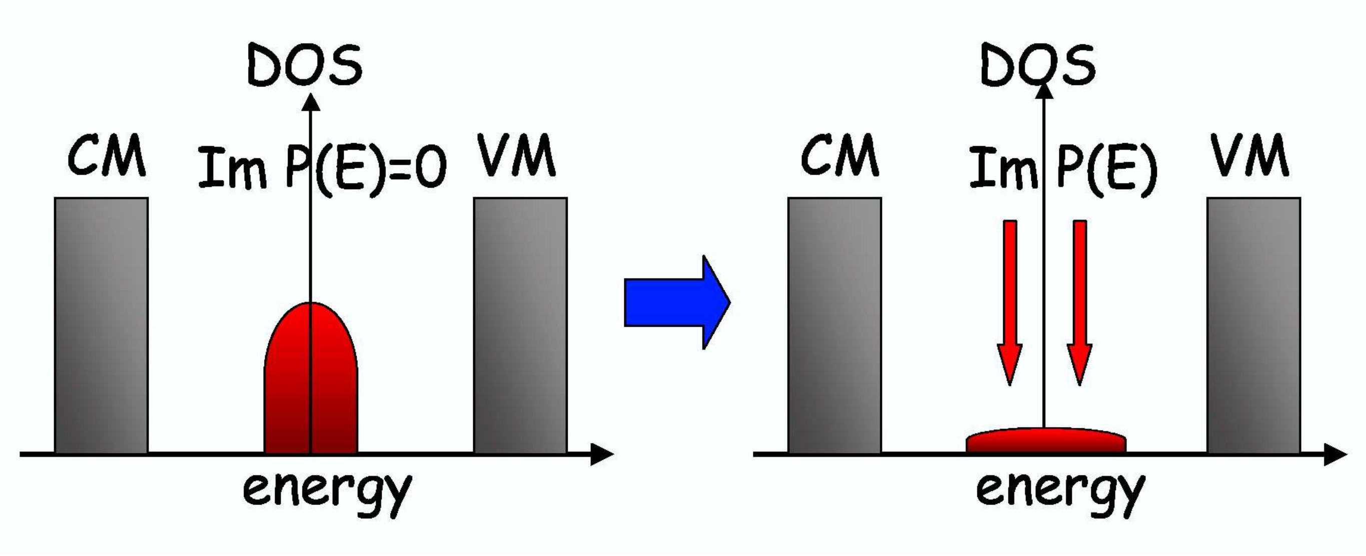
$$+ \underbrace{\sum_j \epsilon_j c_j^\dagger c_j}_{\text{backbone}} - \underbrace{t_\perp \sum_j \left(b_j^\dagger c_j + \mathrm{H.c.} \right)}_{\text{HOMO(LUMO)}-\text{backbone}}$$

$$+ \underbrace{\sum_\alpha \Omega_\alpha B_\alpha^\dagger B_\alpha}_{\text{bath}} + \underbrace{\sum_{\alpha,j} \lambda_\alpha c_j^\dagger c_j \left(B_\alpha + B_\alpha^\dagger \right)}_{\text{bath}-\text{backbone}}$$

$$+ \underbrace{\sum_{\mathbf{k},\alpha=L,R} \epsilon_{\mathbf{k},\alpha} d_{\mathbf{k},\alpha}^\dagger d_{\mathbf{k},\alpha}}_{\text{leads}}$$

$$+ \underbrace{\sum_{\mathbf{k}} t_{\mathbf{k},L} (d_{\mathbf{k},\alpha}^\dagger b_1 + H.c.) + \sum_{\mathbf{k}} t_{\mathbf{k},R} (d_{\mathbf{k},\alpha}^\dagger b_N + H.c.)}_{\text{tunneling terms}}$$

Results (qualitative): Low-bias, strong coupling limit

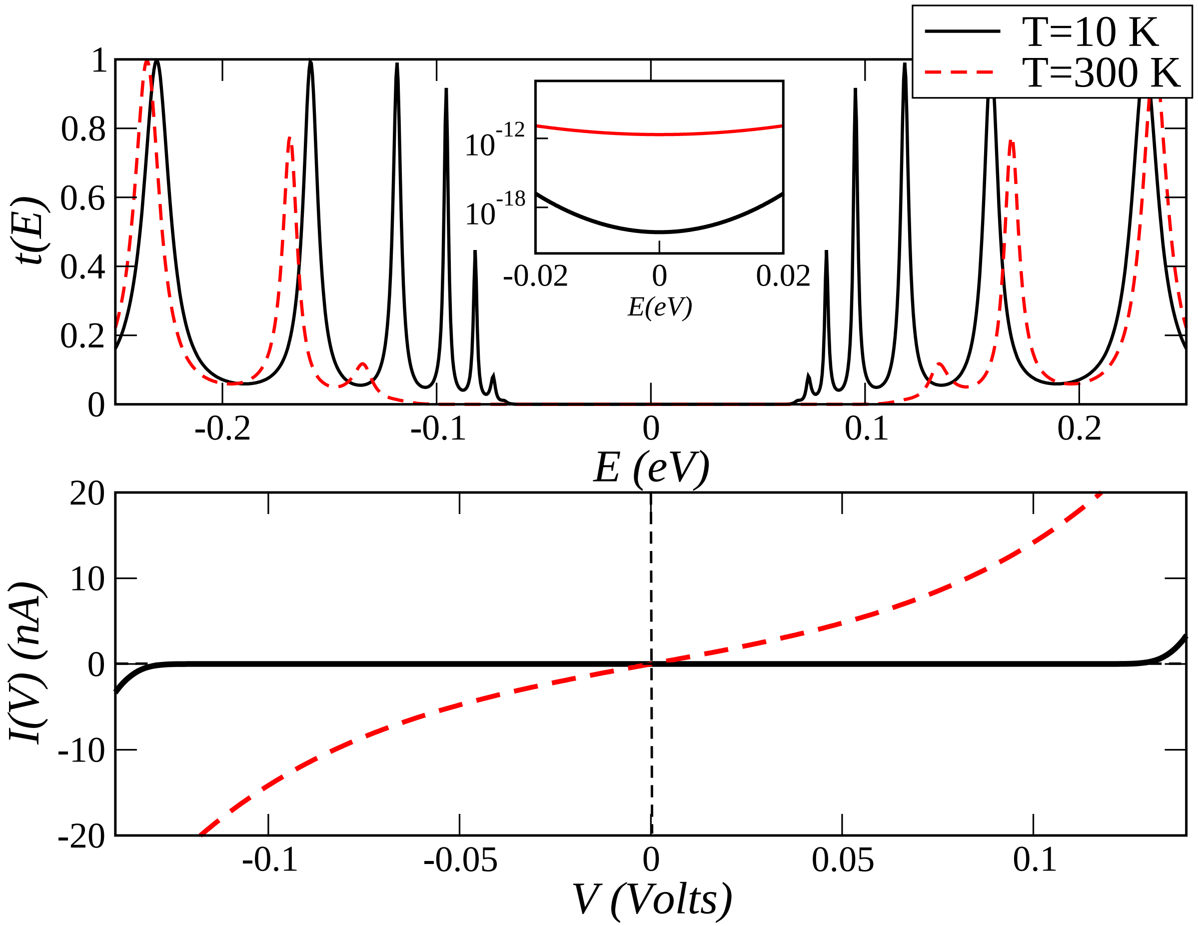


Bath-selfenergy $P(E)$:

Re $P(E)$ $\rightsquigarrow \det|E\mathbf{1} - H - \text{Re}\, P(E)| = 0 \rightsquigarrow$ k_{B}T-dependent manifold

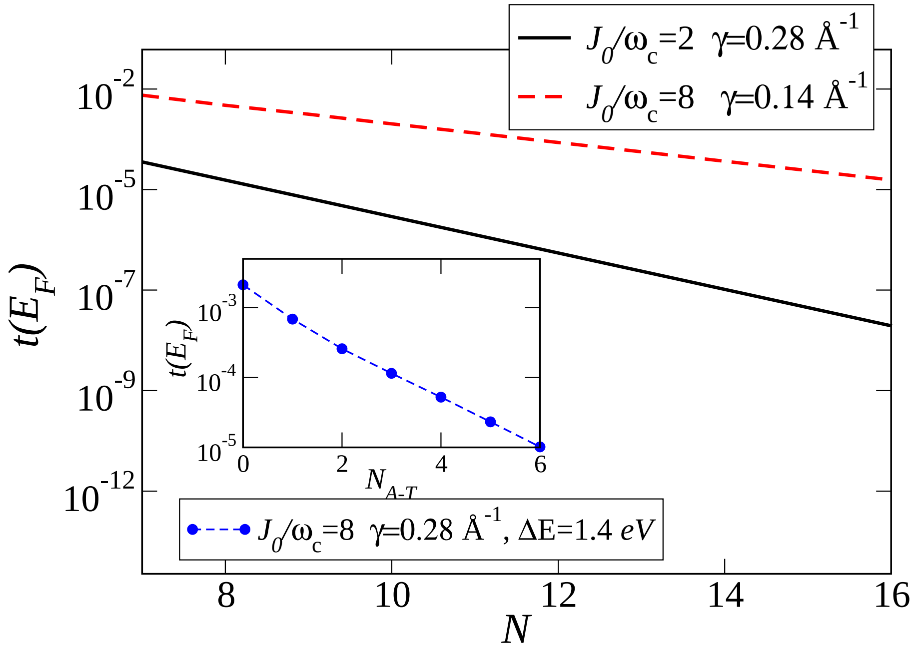
Im $P(E)$ ("friction") $\rightsquigarrow$ incoherent band, pseudo-gap opens

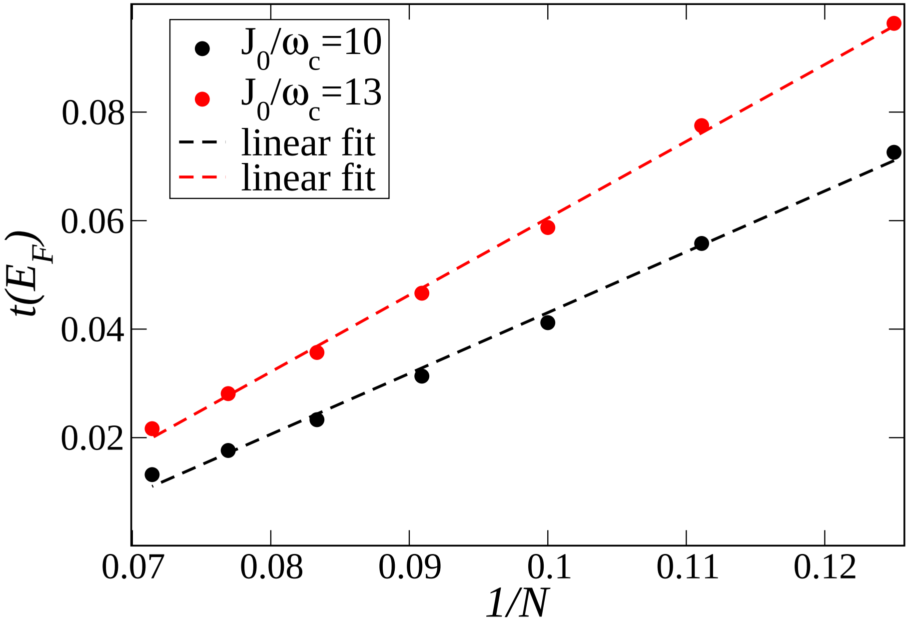
Results: Transmission and low-bias current



Crossover from tunneling (low T) $\rightsquigarrow$ activated (high-T) transport

Results: Scaling of $t(E_F)$ with the chain length $L = Na_{\text{bp}}$ (T=300 K)





- With increasing coupling to the bath transition from

  weak exponential $(\gamma \ll 1)$ $t_F \sim e^{-\gamma L} \Longrightarrow$ algebraic $t_F \sim L^{-\alpha}$

Conclusions and Outlook

- Environment drastically affects charge transport $\rightsquigarrow$

  temperature-dependent (incoherent) DOS near E_F

  $\rightsquigarrow$ non-zero low-bias current at high $k_B T$

  $\rightsquigarrow$ weak exponential or algebraic $L-$dependence

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

- Internal dynamical degrees of freedom? (in progress)

- Sequence complexity ? (in progress)

- Nonequilibrium transport ! (future work)

Green function techniques

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

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

- Green functions ($\hbar = 1$) to $O(t_\perp^2)$

$$G_{jl}(t) = -\mathrm{i}\,\theta(t) < \left\{ b_j(t), b_\ell^\dagger(0) \right\} >$$

$$\mathbf{G}^{-1}(E) = \underbrace{E\mathbf{1} - \mathcal{H}_{\pi-\pi}}_{\mathbf{G}_{\pi-\pi}^{-1}} - \underbrace{\Sigma_L(E) - \Sigma_R(E)}_{\text{leads=WBL}} - \underbrace{t_\perp^2 \mathbf{P}(E)}_{\text{bath-backbone}}$$

$$P_{\ell j}(E) = -\mathrm{i}\,\theta(t) < \left\{ c_\ell(t)\mathcal{X}(t), c_j^\dagger(0)\mathcal{X}(0) \right\} >$$

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

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

Electron-bath correlator in the strong coupling limit

$$t_\perp^2\, P_{\ell j}(z = E + \mathrm{i}\,0^+) = t_\perp^2 \int_0^\infty \mathrm{d}t\, \mathrm{e}^{\mathrm{i}\,zt} \overbrace{G_{\mathsf{c},\ell j}(t)}^{-\mathrm{i}\,[e^{\mathrm{i}\,(\epsilon_{\mathsf{c}}-\Delta)t}]\delta_{\ell j}} \times \overbrace{\langle \mathcal{X}(t)\mathcal{X}^\dagger(0)\rangle_{\mathsf{B}}}^{\mathrm{e}^{-\Phi(t)}}$$

- $J_0/\omega_{\mathsf{c}} > 1 \rightsquigarrow$ short-time expansion $\Phi(t) \approx \mathrm{i}\,t\,\Delta + (\omega_{\mathsf{c}} t)^2\, \kappa_0(T) \rightsquigarrow$ Gaussian integral

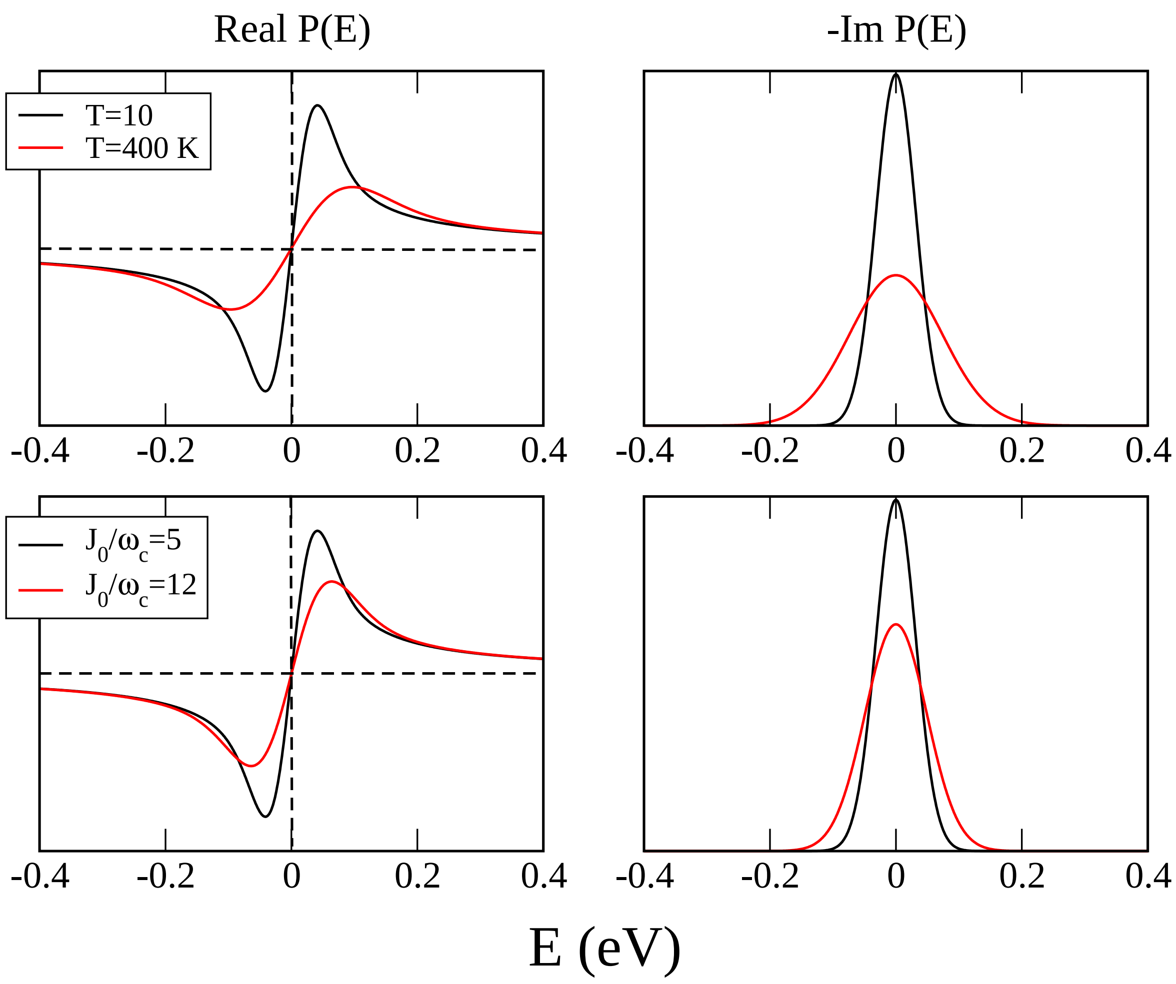
$$t_\perp^2\, P_{\ell j}(z = E + \mathrm{i}\,0^+) = -\mathrm{i}\, t_\perp^2 \delta_{\ell j}\, \sqrt{\pi}\tau_{\mathsf{ph}}\, e^{-\tau_{\mathsf{ph}}^2 z^2} \times \left(1 + \mathrm{erf}\left[\mathrm{i}\,\tau_{\mathsf{ph}} z\right]\right)$$

$$\tau_{\mathsf{ph}}^{-2}(T) = 2\omega_{\mathsf{c}}^2 \kappa_0(T) = \int_0^\infty \mathrm{d}\omega\, J(\omega)\coth\frac{\omega}{2k_{\mathrm{B}}T}$$

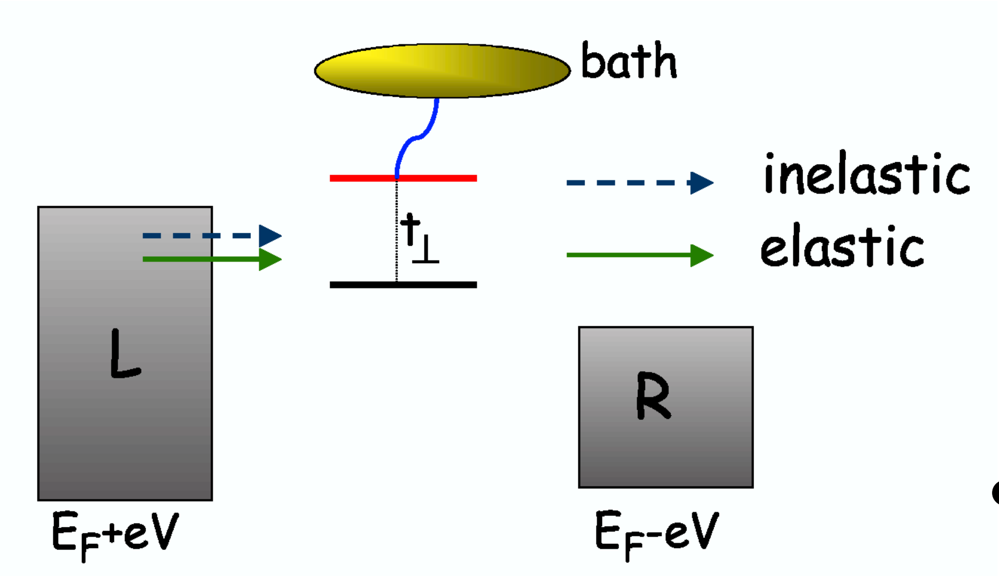
$$\tau_{\mathsf{ph}}(k_{\mathrm{B}}T \ll \omega_{\mathsf{c}}) \sim (J_0\,\omega_{\mathrm{c}})^{-1/2} \qquad \tau_{\mathsf{ph}}(k_{\mathrm{B}}T \gg \omega_{\mathsf{c}}) \sim (k_{\mathrm{B}}T\, J_0)^{-1/2}$$

Behaviour of P$(E, k_B T, J_0/\omega_c)$

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



The electric current



$$I_{\rm L} = \frac{2e}{h}\int dE\,(\Sigma_L^< G^> - \Sigma_L^> G^<)$$

$$I_{\rm L}^{\rm el} = \frac{2e}{h}\int dE\,(f_L - f_R)\,\underbrace{Tr[G^\dagger \gamma_{\rm R} G \gamma_{\rm L}]}_{t(E)}$$

$$I_{\rm L}^{\rm inel} \sim\; t_\perp^2 \times \frac{2e}{h} \times \int\, dE \int\, dE'\, t_\perp^2 \cdots$$

- $-{\rm i}\,\gamma_{\rm L(R)} = \Sigma_{\rm L(R)} - \Sigma_{\rm L(R)}^\dagger$
- $t(E)$ contains full bath dressing !
- for $eV \to 0$ and $O(t_\perp^2) \rightsquigarrow$ neglect $I_{\rm L}^{\rm inel}$

Inelastic current for a dimer

$$j_{\mathrm{L}}^{\mathrm{inel}} = \frac{2e}{h} t_{\perp}^{2} \gamma_{\mathrm{L}} \gamma_{\mathrm{R}} \int dE \int dE' |G_{11}(E)|^2 \underbrace{|G_{21}(E')|^2}_{\sim t_{\perp}^{2}} D_{\mathrm{bath}}^{<}(E - E') \times$$

$$\times [f_{\mathrm{L}}(E)\,(1 - f_{\mathrm{R}}(E')) - (1 - f_{\mathrm{L}}(E'))\,f_{\mathrm{R}}(E)]$$

$$D_{\mathrm{bath}}^{<}(E) = \int dt\, e^{\mathrm{i}\,E\,t} \langle \mathcal{X}(t) \mathcal{X}^{\dagger}(0) \rangle_{\mathsf{B}} = \int dt\, e^{\mathrm{i}\,E\,t} e^{-\Phi(t)}$$